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Rheological characteristics of unstable heavy crude oil-water dispersed mixtures

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ABSTRACT

The rheological characteristics of unstable heavy crude oil-water dispersed mixtures were investigated to improve pipeline transportation. The viscosity of heavy crude oil is 5439 mPa s, and density is 955 kg/m^3 at 30 °C. A series of factors, including oil volume fraction, shear rate, and temperature, was considered systematically to investigate the phenomenon of phase inversion and stability of oil-water mixtures. The results show that the apparent viscosity of a heavy crude oil-water mixture reaches its maximum value at the phase inversion point, and that the non-Newtonian fluid characteristics of unstable heavy crude oil-water mixtures are different than those of fine emulsions or light oil-water mixtures. The addition of emulsifier influences the apparent viscosity and phase inversion point by changing the droplet size distribution. Furthermore, the apparent viscosity of a heavy crude oil-water mixture continues to increase as the median diameter of the dispersed phase decreases. All these are beneficial to the analysis of the rheological characteristics of heavy crude oil-water dispersed mixtures.

1. Introduction

Heavy crude oil and extra heavy crude oil are dense and viscous oils with complex non-Newtonian fluid characteristics, and pipeline transportation remains one of the most efficient and economical methods to transport them despite the difficulties entailed (Oliveira et al., 2018a,b). Global resources of heavy crude oil are at least of the conventional light crude oil, and this provides an interesting situation with respect to the economics of petroleum development (Martinez-Palou et al., 2011). Crude oil is always present as a mixture with water because of the presence of water in oil wells or the injection of water into oil wells to increase oil production (Kelesoglu et al., 2012; Akbari and Nour, 2018). Heavy crude oil-water mixtures are easily formed as dispersed mixtures owing to shearing or stirring forces from valves, pumps, elbows, etc. Simultaneously, the existence of certain amounts of surface-active components of heavy crude oil, such as asphaltenes and resins, enhances dispersion (Mironova and Ilyin, 2018; Oliveira et al., 2018a,b). Unfortunately, heavy crude oil-water mixtures are unstable unlike normal oil-water emulsions, and hence exhibit different non-Newtonian fluid behaviors than the latter even at identical oil volume fractions (Kelesoglu et al., 2012; Zhang et al., 2013). The complex rheological characteristics affect pipeline flow behaviors and parameters, rendering it significantly difficult to design two-phase oil-water transportation and other petroleum exploitation systems (Masalova et al., 2003; Plasencia et al., 2013; Sun et al., 2018). Therefore, the rheological characteristics of heavy crude oil and its mixtures, especially unstable mixtures characteristic of the petroleum industry, must be understood to improve oil field development and pipeline transportation.

Significant research has been conducted on the rheological characteristics of pure crude oil and crude oil-water emulsions with different areas of focus, including apparent viscosity prediction and stability analysis (Aomari et al., 1998; Li et al., 2014; Wen et al., 2016). The rheological characteristics of oil-water emulsions are affected by several parameters and can be presented as functions of viscosity of dispersed phase, volume fraction of dispersed phase, viscosity of continuous phase, shear rate, droplet size distribution, temperature, etc. Over the past century, a few correlations and models (empirical or semi-empirical), as summarized by Zhang et al. (2013), have been developed for predicting the apparent viscosities of oil-water emulsions. However, most models are based on light oil and fine emulsions with low dispersed phase fractions that always exhibit Newtonian fluid behavior. It has been established recently that heavy crude oil-water emulsions or coarse oil-water emulsions can exhibit non-Newtonian behavior even at low dispersed phase fractions, and that the apparent viscosities of crude oil-water mixtures are greater than those of

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corresponding stable emulsions (Neto et al., 2019). Research (Meriem-Benziane et al., 2012; Zhang et al., 2013) has shown that in the case of crude oil-water emulsions, the apparent viscosity varies with oil content and attains peak value at the phase inversion point. Also, their rheological behaviors can be described by the Ostwald de Waele or Herschel-Bulkley models at different oil concentrations. A study of the effects of temperature on the apparent viscosities of oil-water emulsions has revealed that viscosity decreases with increase in temperature, and the authors prescribed the Arrhenius model to describe the relation between the two parameters (Kolotova et al., 2018). Based on existing prediction models and experimental results, a few new prediction models have been developed to calculate the relative viscosities of crude oil-water emulsions more accurately (Equation (1), Zhang and Xu, 2019, Equation (2), Carmo et al., 2019). The effect of droplet size distribution on apparent viscosity was first considered by Zhang and Xu, 2019, who established that viscosity increased as mean diameter decreased in their experimental region.

$$\mu_r = {}^{\mu_a}\!/_{\mu_c} = \exp\left(\frac{2.5\varepsilon_d}{1 - k_e\varepsilon_d}\right)\left(\frac{\mu_d}{\mu_c}\right)^h \tag{1}$$

$$\mu_r = {}^{\mu_a}\!/_{\mu_c} = E_\delta\left[k_1\left(\frac{\varepsilon_d}{1 - k_2\varepsilon_d}\right)^\lambda\right] \tag{2}$$

Here, μ_a is the apparent viscosity, μ_c is the viscosity of the continuous phase, μ_d is the viscosity of the dispersed phase, ε_d is the volume fraction of the dispersed phase, and E_δ is the Mittag-Leffler function.

The stability of crude oil-water mixtures has been researched in several instances, but the effects of stability on the rheological characteristics of the mixtures have not been studied sufficiently. Surfactants, such as the natural components of crude oil and additive emulsifiers, have been experimentally found to increase the stability of crude oil-water emulsions, and the phenomenon of separation has always been used to describe the degree of stability (Kang et al., 2012). Furthermore, the speed and duration of mixing significantly influence the stability of oil-water mixtures (Ahmed et al., 1999). Microstructure and mean diameter have been used to analyze stability in the literature (Wong et al., 2015, 2018). In the case of unstable oil-water mixtures, Feng et al. (2019) studied the viscoelastic characteristics of heavy crude oil-water dispersed mixtures. However, no other rheological characteristic of unstable heavy crude oil-water dispersed mixtures has been researched systematically although such mixtures are different than stable emulsions and influence the design of crude oil exploitation and transportation systems (Chen and Tao, 2005). Therefore, it is necessary to study the rheological characteristics of unstable heavy crude oil-water mixtures, characteristic of the petroleum industry, to improve two-phase pipeline flow.

In this work, the rheological characteristics of unstable heavy crude oil-water dispersed mixtures were investigated using a rotational rheometer. The oil volume fraction was in the range 0–1, which includes the continuous phase inversion region. A series of factors, including oil volume fraction, shear rate, and temperature, was considered systematically. The oil-water mixtures were prepared by adding emulsifier and employing different stirring parameters. The stabilities of the mixtures were investigated based on the microstructures obtained using a microscope and charge-coupled device (CCD) system, and the influence of stability on the rheological characteristics of the mixtures was additionally investigated.

2. Experimental

2.1. Materials

The heavy crude oil used in this work was obtained from the Bohai oil field in Liaodong Bay in China. The density of the heavy crude oil is 955 kg/m^3 and shear viscosity is 5439 mPa s at 30 °C. Tap water, with a density of 1000 kg/m3, is used as the other phase. And, the degree of mineralization of the tap water is 245 mg/L. The weight fractions of saturates, aromatics, resins, and asphaltenes (SARA) obtained from the SARA analysis of the heavy crude oil are 33.2%, 26.2%, 37.2%, and 3.4%, respectively. The effects of temperature on the density and viscosity of the heavy crude oil are displayed in Fig. 1, which shows that both density and viscosity continued to decrease with increase in temperature. Other rheological characteristics of pure heavy crude oil, including shear thinning, thixotropy, and yield stress, can be found in the work of Zhang et al. (2017).

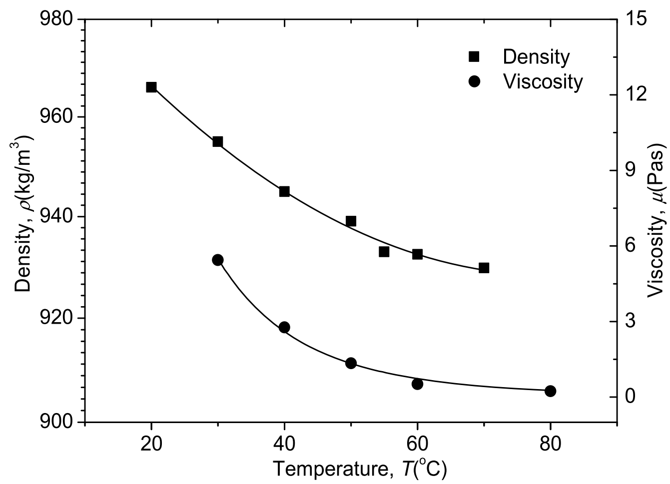


Fig. 1. Physical properties of the heavy crude oil sample at different temperatures.

2.2. Preparation of dispersed mixtures

In this study, a series of stable and unstable two-phase oil-water dispersed mixtures was prepared to consider the effects of the dispersed phase distribution and different stability characteristics. The definition of the stable mixtures can be found in the reference (Wong et al., 2015). The mixtures had different oil volume fractions in the range 0–1. A three-blade stirrer was used to homogenize the two-phase oil-water solutions at a fixed speed of 1000 rpm. Furthermore, different stirring times of 100s, 300s, 600s, and 1200s were chosen to obtain the series of experimental mixtures with different dispersed phase distributions and stability characteristics. The two-phase oil-water mixtures grew more stable with increase in stirring time. Sodium dodecyl benzene sulfonate (SDBS), supplied by Sinopharm Chemical Reagent Co. Ltd. (China), was chosen as the emulsifying agent to obtain heavy crude oil-water emulsions of different stabilities.

The microstructure of each two-phase oil-water dispersed mixture prepared was obtained using a trinocular optical microscope Olympas BX43 with an adapted CCD camera ProgRes C5. Subsequently, the dispersed phase distribution was measured using the relevant microstructure images. The amounts of water precipitated from the mixtures at different settling time after preparation was measured, and the results were used to quantitatively measure the stability characteristics of the mixtures.

2.3. Rheological measurements and procedure

The rheological characteristics of pure crude oil and its mixtures were measured using the rotational rheometer HAKKE RS6000, and an assorted coaxial cylinder sensor system Z38 was employed in all experiments. The diameter of the rotor is 38 mm, and the gap width between the rotor Z38 and cup Z43 is 2.5 mm, which is sufficiently large compared to the droplet diameter of the dispersed phase. The shear rate and torque of the rheometer are in the ranges 0.001–1500s^{-1}, and

0.05 μNm-200 mNm, respectively. Circulating water bath temperature control units were available to handle experimental temperatures in the range 0–100 °C with an accuracy of 0.1 °C.

In this study, the control rate model of the rheometer, wherein the range of shear rate was designed and shear stress was measured simultaneously, was used to measure the rheological characteristics of the heavy crude oil-water dispersed mixtures. The experimental temperature was in the range 20–80 °C, which includes all the typical operating temperatures employed in the petroleum industry. Furthermore, the duration of measurement of rheological characteristics was controlled to ensure the unstable heavy crude oil-water dispersed mixtures remained in quasi-stable status during the measurements. Additionally, the stability conditions of the mixtures were monitored using a high-speed camera, according to the method described by Feng et al. (2019). All experiments were repeated at least thrice to ensure accuracy of results.

3. Results and discussion

It is known that oil-water emulsions always exhibit the characteristics of non-Newtonian fluids with higher dispersed-phase volume fractions, and that volume fraction has significant influence on the degree of non-Newtonian behavior (Dou and Gong, 2006; Meriem-Benziane et al., 2012; Zhang et al., 2013). According to research, the Ostwald de Waele model exhibits great accuracy when used to discuss the non-Newtonian behavior of oil-water emulsions. Hence, the model was chosen to analyze the experimental results of unstable heavy crude oil-water dispersed mixtures in the present study. The Ostwald de Waele model for non-Newtonian fluids is based on two parameters, as described in the following equation.

$$\tau = k(\gamma')^n \tag{3}$$

Here, τ is the shear stress in Pa, γ' is the shear rate in s^{-1}, k is the consistency index in Pa· s^n, and n is the flow behavior index.

3.1. Stability analysis of heavy crude oil-water mixtures prepared

To investigate the influence of stability on rheological characteristics, a series of heavy crude oil-water dispersed mixtures of different degrees of stability was prepared by employing different stirring durations. Four stirring times, i.e., 100s, 300s, 600s, and 1200s were chosen, and the stirring speed was controlled at 1000 rpm in all cases. The phenomenon of separation was used to measure the stabilities of the oil-water mixtures, and their micro-structures were obtained simultaneously. Here, the separated water ratio, α, as defined in Equation (4), was introduced to analyze the stabilities of the mixtures. The water separated (V_{ws}) is the volume of water separated from the unstable oil-water dispersed mixture after stirring, and water content (V_{wc}) is the total volume of water present in the mixture.

$$\alpha = \frac{water\ separated,\quad V_{ws}(\mathrm{mL})}{water\ content,\quad V_{wc}(\mathrm{mL})} \tag{4}$$

Fig. 2 displays the phenomenon of separation in the heavy crude oil-water dispersed mixtures prepared, and several settling durations were chosen for the purpose of comparison. The oil volume fraction is 0.5, and the stirring times are 100s and 300s. The figure shows that both the oil-water mixtures maintained stable status for the first 5 min, and this is deduced from the lack of appearance of separated water. Here, this status is defined as quasi-stable because this condition cannot be maintained throughout the study. Subsequently, separated water appeared at the bottom as settling time increased. It can be seen that for a given settling time, the volume of separated water is lower when the stirring time of preparation is longer. This implies that the heavy crude oil-water dispersed mixtures prepared are more stable when longer stirring time is employed.

The variations in the separated water ratios as functions of settling time of the oil-water mixtures prepared are given in Fig. 3. It shows that the heavy crude oil-water mixture with oil volume fraction of 0.5 and stirring time of 100s was the most unstable, and that the oil and water in this mixture separated completely after a settling duration of 180 min. Furthermore, the addition of SDBS can greatly increase the stability of crude oil-water dispersed mixtures, and this can be determined by comparing mixtures prepared with and without SDBS (Channam, 2005).

The effect of stability on the rheological characteristics of heavy crude oil-water dispersed mixture was researched. Fig. 4 gives the relations between apparent viscosity and shear rate in the case of four oil-water mixtures prepared under different stirring times without emulsifier. The oil volume fraction was 0.5. Firstly, the mean diameters of the four mixtures were measured by analyzing the relevant micro-structures. The results are displayed in Table 1, the mean diameters were 42.0 μm, 34.5 μm, 22.1 μm, and 17.0 μm of the mixtures prepared under stirring times of 100s, 300s, 600s, and 1200s, respectively. The mean diameter decreased as stirring time increased. Simultaneously, the apparent viscosity increased as mean diameter decreased within our experimental range, as can be seen from Fig. 4 and Table 1. Therefore, droplet size distribution, which has not received adequate attention in existing prediction models, must be considered in the calculation of the apparent viscosities of unstable heavy crude oil-water dispersed mixtures. Moreover, we can see that unstable heavy crude oil-water dispersed mixtures always exhibit the characteristics of non-Newtonian fluids, and their apparent viscosities decreased as shear rate increased, as seen in Fig. 4.

3.2. Rheological characteristics of mixtures with different phase fractions

The stirring time was fixed at 300s to obtain unstable heavy crude oil-water dispersed mixtures with identical stabilities. The oil volume fraction was in the range 0–1, and the emulsifier SDBS was used to prepare mixtures with greater stability. The rheological characteristics of all the mixtures were measured under the same conditions. More importantly, the rheological characteristics of the mixtures near the continuous phase inversion points were studied by combining their relevant microstructures.

The variations in apparent viscosity with shear rate in the unstable heavy crude oil-water dispersed mixtures are displayed in Fig. 5. In the case of mixtures without emulsifier, apparent viscosity is higher in mixtures with oil volume fraction greater than 0.5 than in mixtures with oil volume fraction lower than 0.4, as seen in Fig. 5(a). Under the preparation conditions in this study, mixtures with oil volume fraction greater than 0.5 presented the water-in-oil form, and those with oil volume fraction less than 0.4 displayed an oil-in-water form. Where the oil-water mixtures changed from the water continuous to the oil continuous was defined as the phase inversion phenomenon. The oil volume fraction in the range 0.4–0.5 was defined as the transition area wherein the water continuous and oil continuous flow patterns occurred simultaneously. The phase inversion point should be influenced by the physical parameters of the two phases and the prepared conditions greatly (Salager et al., 1983). In the heavy crude oil-water dispersed mixtures prepared with the addition of SDBS, the transition area was in the range 0.5–0.6. Thus, the addition of SDBS in the preparation of oil-water mixtures evidently affected their phase inversion points where emulsifiers are always expected to stabilize more water in oil.

To analyze the non-Newtonian fluid behavior of unstable heavy crude oil-water dispersed mixtures, the Ostwald de Waele model was chosen to fit the experimental results. Table 2 gives the fitting parameters of the consistency index, k, and flow behavior index, n, in this model. The parameter R^2 is the correlation coefficient, which is used to measure the fitting consequence. It shows that the value of n was greater than 1 for all oil-in-water mixtures, with or without emulsifier, and smaller than 1 for water-in-oil mixtures. Therefore, the oil-in-water dispersed mixtures displayed varying degrees of shear thickening fluid

Fig. 2. Monitoring separation of the oil-water mixtures prepared without emulsifier (oil volume fraction = 0.5).

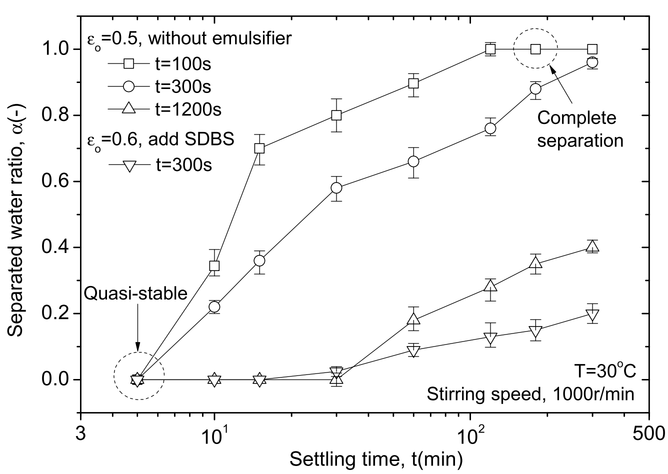


Fig. 3. Variations in separated water ratio as functions of settling times of the oil-water mixtures prepared.

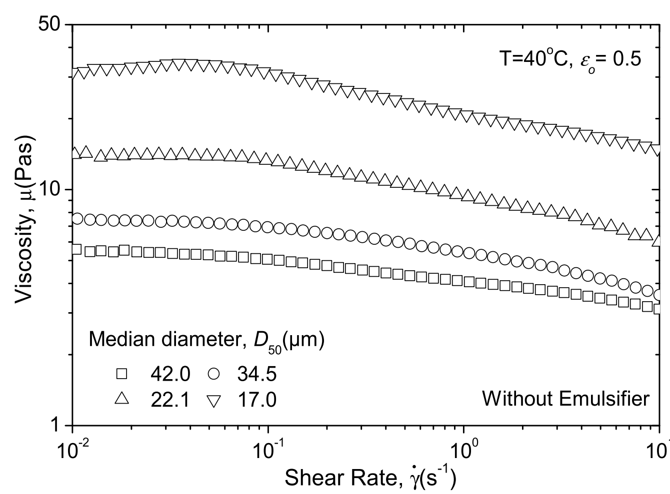


Fig. 4. The relation between apparent viscosity and shear rate of mixtures with different median diameters (oil volume fraction, 0.5).

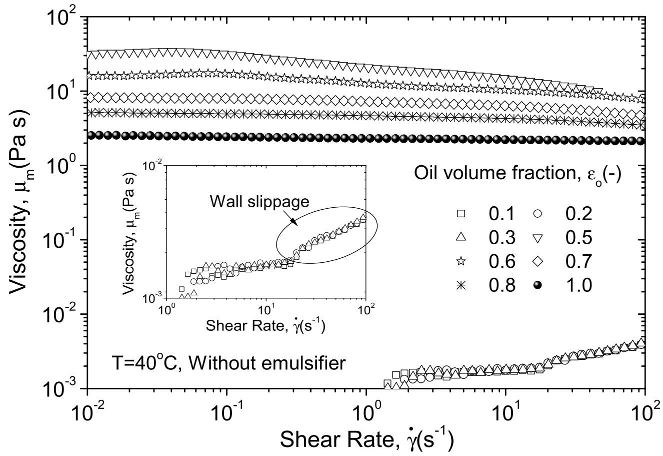
behavior, while the water-in-oil dispersed mixtures inversely displayed different degrees of shear shinning fluid behavior. Furthermore, the addition of SDBS cannot affect the non-Newtonian fluid behavior of the oil-water mixtures prepared but can influence their degrees of shear thickening or shear thinning.

Fig. 6 displays the variations in the relative viscosities of the unstable heavy crude oil-water dispersed mixtures with increase in oil volume fraction at the same shear rate and temperature. The relative viscosity initially increased and reached the maximum value near the phase inversion point, following which it decreased to 1, which is the viscosity of pure oil. The effect of emulsifier on relative viscosity was greater in the case of oil-in-water mixtures than in water-in-oil mixtures. The addition of SDBS always increases the relative viscosity of oil-in-water mixtures by influencing the microstructure. And, the emulsifier has a greater influence on the microstructure of the oil-in-water mixtures than the water-in-oil mixtures where the dispersity and diameter of the dispersed phase oil in oil-in-water mixtures will be changed greater than the continuous phase water in the water-in-oil mixtures.

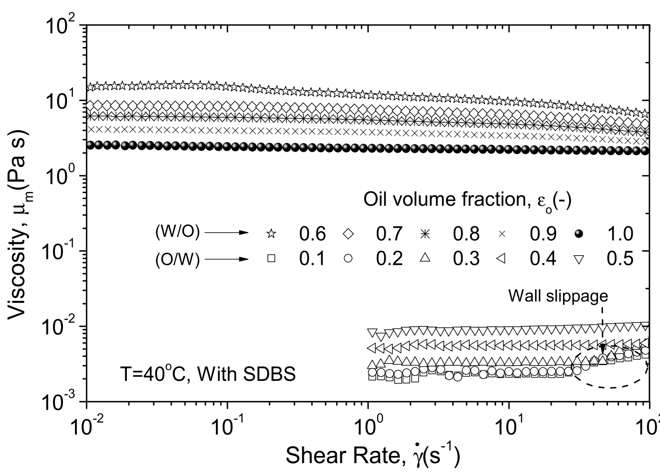
Table 1
Median diameter and rheological analysis of heavy crude oil-water mixtures under different conditions of preparation (oil volume fraction, 0.5).

Stirring time t(s)	Median diameter D_{50} (μm)	Apparent viscosity ($\gamma' = 1s^{-1}$), μ_a(mPa.s)
100	42.0	4114
300	34.5	5468
600	22.1	9515
1200	17.0	21202

The microstructures of the heavy crude oil-water dispersed mixtures



(a) No emulsifier added



(b) SDBS added as emulsifier

Fig. 5. Rheological characteristics of unstable heavy crude oil-water mixtures of different oil volume fractions.

Table 2
Fitting parameters of Ostwald de Waele model for heavy crude oil-water dispersed mixtures with different oil volume fractions (T = 40 °C).

ε_o (−)	Without emulsifier			With SDBS		
	k	n	R^2	k	n	R^2
0.1	0.0012	1.1531	0.9941	0.0022	1.0172	0.9989
0.2	0.0012	1.1928	0.997	0.0023	1.0318	0.9981
0.3	0.0012	1.1764	0.9975	0.0031	1.0283	0.9998
0.5	24.7627	0.7693	0.9997	0.0076	1.0694	0.9999
0.6	14.4117	0.8657	0.9998	14.5178	0.8299	0.9997
0.7	8.8192	0.8636	0.9998	8.8389	0.8678	0.9999
0.8	5.3872	0.9058	0.9999	6.4443	0.8834	0.9999

prepared are shown in Fig. 7. The mixtures, when prepared without emulsifier, were in the oil-in-water form when the oil volume fraction was less than 0.4; when the mixtures were prepared with SDBS, the critical oil volume fraction was 0.5. Moreover, the oil-water mixture, prepared without emulsifier, with oil volume fraction of 0.5 displayed the water-in-oil form, and this can be used to prove the effects of the relevant rheological characteristics. Thus, the effects of emulsifier and phase inversion point on rheological behavior must be considered systematically.

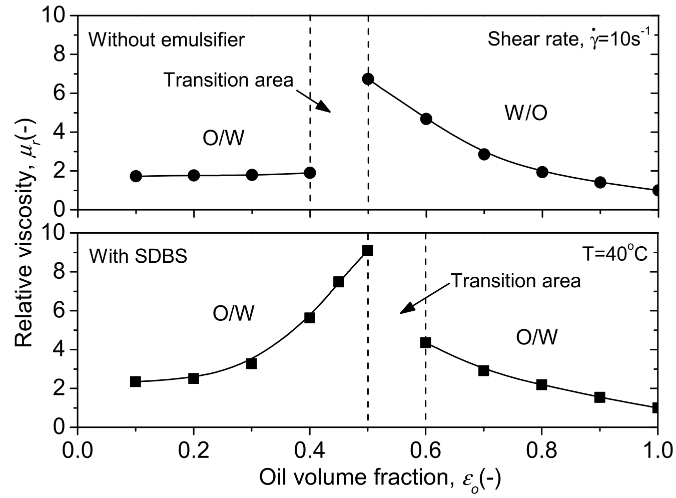


Fig. 6. Relative viscosities of oil-water dispersed mixtures as functions of oil volume fraction (T = 40 °C; γ' = 10s^{-1}).

3.3. Effect of temperature on apparent viscosity

Temperature significantly influences fluid viscosity, and this relationship is governed by the Arrhenius model (Equation (5)). In this section, the effect of temperature on the apparent viscosities of the unstable heavy crude oil-water dispersed mixtures is studied.

$$\mu_a = A \times e^{B/T} \tag{5}$$

Here, A and B are constants dependent on both system and shear rate. T is the absolute temperature in °C.

The effects of temperature on the apparent viscosities of two kinds of heavy crude oil-water dispersed mixtures are displayed in Fig. 8. The apparent viscosities of pure heavy crude oil and all its mixtures decreased exponentially with increase in temperature. The Arrhenius model can be used to accurately describe the relationship, which displays the same trend in the case of normal oil-water emulsions.

3.4. Relative viscosity prediction and analysis using existing models

Several prediction models based on different theories are available in the literature for the calculation of the apparent viscosities of oil-water dispersed mixtures. The stabilities and non-Newtonian fluid characteristics of oil-water dispersed mixtures were considered in addition to a comparison with fine emulsions. In the present study, two new prediction models (Equation (1), Zhang and Xu, 2019, Equation (2), Carmo et al., 2019) were chosen to analyze the apparent viscosities of the unstable heavy crude oil-water dispersed mixtures. The average deviation was introduced to calculate the prediction accuracy of these models.

$$\theta = \frac{1}{n}\sum_{i=1}^{n}\left|\frac{\mu_{rp}-\mu_{re}}{\mu_{re}}\right| \times 100 \tag{6}$$

Here, θ is the average deviation (%), μ_{rp} is the predicted relative viscosity, and μ_{re} is the experimental relative viscosity.

Fig. 9 displays a comparison between the experimental and calculated values of relative viscosity. In the literature, the model of Carmo et al. (2019) has been used to calculate relative viscosity only in the case of water-in-oil mixtures. This shows that the chosen models can predict the relative viscosities of the unstable heavy crude oil-water dispersed mixtures in this work accurately. However, the phase inversion point first needs to be determined, and the degrees of stability of the mixtures prepared should be considered simultaneously. Therefore, the apparent viscosity of heavy crude oil-water dispersed mixtures can be predicted by through the understanding of the phase distribution

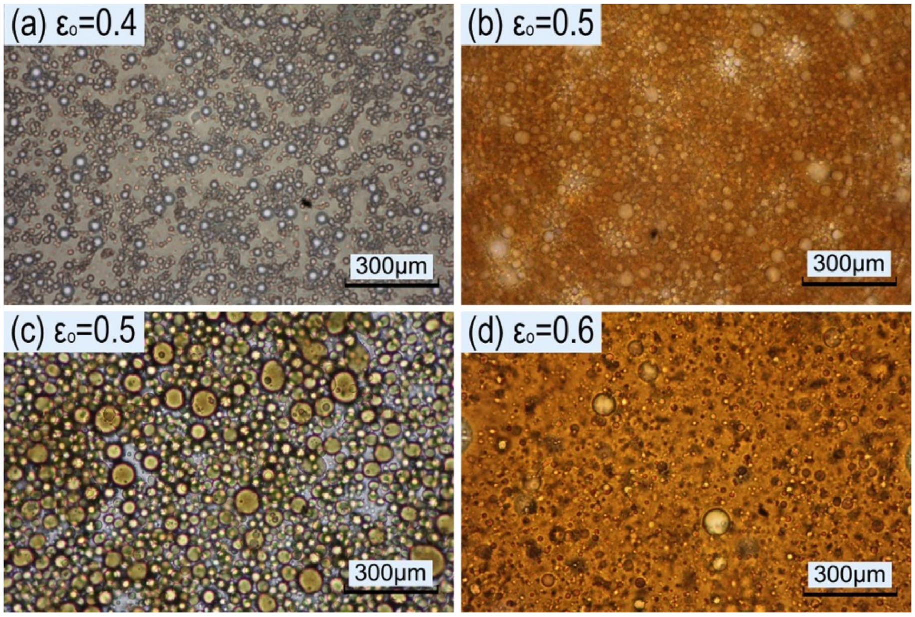


Fig. 7. Microstructures of the prepared oil-water mixtures near the phase inversion point (a and b, without emulsifier; c and d, with SDBS).

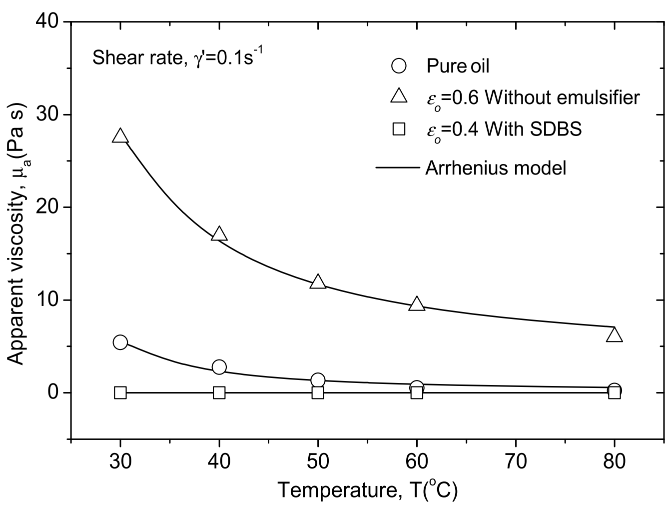


Fig. 8. Effect of temperature on apparent viscosities of pure heavy crude oil and its mixtures.

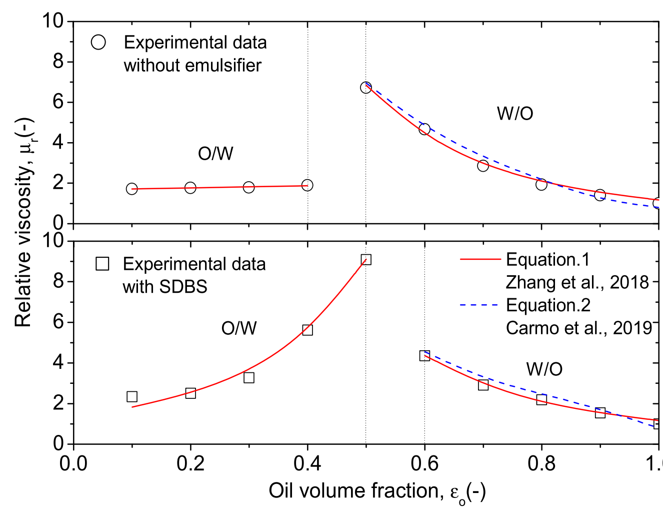


Fig. 9. Relative viscosity prediction by two new models in the literature (T = 40 °C; γ' = 10s^{-1}).

and these prediction models. The average deviation is given in Fig. 10. The predicted values of relative viscosity coincided greatly with the experimental data. The average deviations of the values from Equation (1) and Equation (2) were 6.58% and 6.81%, respectively. It is great useful to calculate the pressure drop of the heavy crude oil-water dispersed flow in pipeline.

4. Conclusion

The rheological characteristics of unstable heavy crude oil-water dispersed mixtures were investigated to improve pipeline transportation in the petroleum industry. A series of factors, including oil volume fraction, shear rate, and temperature, was considered, and the effects of the phenomenon of phase inversion and stability of the mixtures on their rheological characteristics were studied emphatically.

The apparent viscosity values of the heavy crude oil-water mixtures varied with oil volume fractions and reached their maximum values at the phase inversion points. Unlike fine emulsions and light oil-water dispersed mixtures, all mixtures in the present study exhibited non-Newtonian fluid characteristics even at low dispersed-phase fractions. The Ostwald de Waele model can be used to describe the behavior of non-Newtonian fluids accurately. The addition of emulsifier influenced the apparent viscosity and phase inversion point by changing the droplet size distribution. The apparent viscosities of heavy crude oil-water mixtures with different stabilities are vastly different, apparent viscosity continued to increase as the median diameter of dispersed phase decreased in our experimental conditions.

The effect of temperature on apparent viscosity can be described by the Arrhenius model wherein an increase in temperature causes a reduction in the apparent viscosity of an unstable heavy crude oil-water dispersed mixture. This trend is similar to that observed in fine oil-water emulsions. The apparent viscosity of an unstable heavy crude oil-water dispersed mixture can be calculated accurately using the prediction models of Zhang and Xu, 2019 and Carmo et al. (2019). However, the phase inversion point needs to be determined first, and the degree of stability of the mixtures prepared ought to be considered simultaneously.

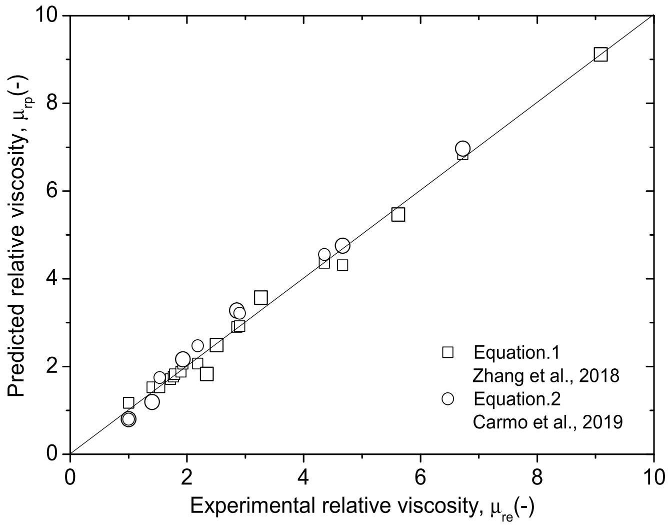


Fig. 10. Prediction accuracy of existing models for heavy crude oil-water dispersed mixtures.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.petrol.2019.106299.

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