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#### Viscoelastic characteristics of heavy crude-oil-water two-phase dispersed mixtures

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#### Abstract

The viscoelastic characteristics of unstable heavy crude-oil-water dispersed mixtures are investigated to improve pipeline transportation. The effects of temperature, dispersed phase volume fraction, and droplet size distribution are considered on experiments. And, the unstable characteristics of heavy crude-oil-water dispersed mixtures, which display the similar droplet size distribution as the two-phase pipeline flow, is emphasized in this study. As a result, the mixing of water into heavy crude oil does not considerably affect the storage modulus that maintains a constant value even with the changing oil volume fractions. The loss modulus increases with the increased oil volume fractions of the oil-in-water dispersed mixtures. It increases to its maximum value when the oil volume fraction increases to the phase inversion point of oil-water mixtures. After the phase inversion point, the loss modulus decreases as the oil volume fraction increases which can be described by the modified Mooney model accurately. The viscoelastic parameters decrease with increasing temperature because the mechanical strength of the interfacial film and the ability to resist droplet deformation due to shear force decrease when the temperature increases. The storage modulus, loss modulus, and complex viscosity increase as the median diameter decreasing with the approximate exponent behaviors. This is because the link force between two droplets is increased by a smaller distance through a longer stirring time which will decrease the median diameter. Also, the smaller droplet, which is more difficult to deform by shear stress, can increase the values of the viscoelastic parameters. Furthermore, the critical elastic dominant

and viscous dominant are investigated through the critical frequency. In conclusion, the unstable heavy crude-oil-water dispersed mixture displays the great different viscoelastic characteristics compared to the normal crude-oil-water emulsions. The unstable characteristics must be considered in the design of the heavy crude oil and water two-phase dispersed flow system. It is important for understanding the pipeline transportation system of heavy crude-oil-water mixtures.

Key words: heavy crude-oil-water mixture, unstable, viscoelasticity, phase fraction, droplet size distribution.

#### 1. Introduction

Heavy crude oil is any type of crude oil that does not flow easily with an API value of less than 20 and high viscosity. The resources of heavy crude oil are more than twice than those of conventional light crude oil worldwide; moreover, the mining of heavy crude oil is becoming more important to supply the fuel for economic development. However, owing to the different components of saturates, aromatics, resins, and asphaltenes, heavy crude oil can demonstrate nearly complex rheological behaviors compared to light crude oil, such as shear thinning, thixotropy, yield stress, and viscoelasticity (Liu et al., 2018; Ilyin et al., 2016). The complex rheological characteristics must significantly affect the production of heavy crude oil (Kane et al., 2004; Sierra et al., 2016). Meanwhile, as a normal operation practice in the oil field, oil and water are always transported together in a pipeline owing to the oil reservoir conditions and economic considerations. The drastic shearing and mixing effects, including flowing, pumps, and valves, can cause dispersion from one phase to another. The mixing of water will increase the complexity of the rheological characteristics of heavy crude oil because of the interaction between the dispersed droplets and the interface effect of the dispersed and continuous phases (Pal, 2000; D'Avino and Maffettone, 2015). And, the crude-oil-water dispersed

mixtures display the unstable characteristics which will separate after shearing and show the great different microstructures of normal stable emulsions. It will significantly increase the difficulties in the design of pipeline transportation system (Kelesoglu et al., 2015). Therefore, it is important to understand the rheological characteristics of the heavy crude-oil-water dispersed mixtures.

The viscoelastic characteristic of the heavy crude oil and water two-phase dispersion mixture is an important rheological characteristic (Meriem-Benziane et al., 2012). In particular, it affects the shutdown and restart sections of the pipeline transportation when fluid deformation occurs before reaching the yield stress. The fluid will restore its initial state because of the elastic characteristic if the stress is removed and less than the yield stress. To research the viscoelastic characteristic, the physical analogy method was introduced to describe the relation between stress and deformation (Mendes and Thompson, 2012; 2013). The spring and dashpot models are used in these physical analogy models. Additionally, the Maxwell model (Eq. 1) (Maxwell, 1873) and Kelvin-Voigt (Eq. 2) (Palierne, 1990) model are two classical linear-viscoelastic models obtained from the series and parallel connections of a spring and dashpot, respectively.

$$\tau_{ij} + \frac{\eta}{G} \frac{\partial \tau_{ij}}{\partial t} = \eta \frac{\partial \gamma_{ij}}{\partial t}$$
(1)

$$\tau_{ij} = G\gamma_{ij} + \eta \frac{\partial \gamma_{ij}}{\partial t} \tag{2}$$

where  $\eta$  is the viscosity of dashpot in Pa·s; and G is the elastic modulus of the spring in Pa.

Currently, the small amplitude oscillation measurement is the primary method to measure and analyze the linear viscoelastic characteristics of fluid. The stress is assigned with an amplitude  $\tau_0$  and an angular velocity  $\omega$  in a CS test model of a rotational rheometer. The corresponding strain is measured with the strain amplitude  $\gamma_0$  and the phase angle  $\delta$ . The parameter complex modulus  $G^* = \tau/\gamma$  $= G' + i^*G''$  is introduced to describe the viscoelastic characteristic, where G' is the storage modulus used to describe the elastic characteristic of fluid, G'' is the loss modulus used to describe the viscosity characteristic of fluid, and i is the complex number. In this study, the small amplitude oscillation measurement was used to investigate the viscoelastic characteristics of the heavy crude-oil-water dispersed mixtures.

Study on the viscoelastic characteristics of oil-water dispersed mixtures was performed on a series of aspects involving micro-theory, the effects of dispersed phase fraction, and some other measurement conditions. Palierne (1990) demonstrated a model to predict the complex modulus of the single droplet size emulsions in the linear viscoelastic region:

$$G^* = G_c^* \left[ \frac{1+3/2H\varphi_d}{1-H\varphi_d} \right] \tag{3}$$

where  $G_c^*$  is the complex modulus of the continuous phase;  $\varphi_d$  is the dispersed phase volume fraction.

$$H = \frac{2[(G_d^* - G_c^*)(19G_d^* + 16G_c^*) + (4\sigma/R)(5G_d^* + 2G_c^*)]}{(2G_d^* + 3G_c^*)(19G_d^* + 16G_c^*) + (40\sigma/R)(G_d^* + G_c^*)},$$
(4)

where  $G_d^*$  is the complex modulus of the dispersed phase,  $\sigma$  is the interfacial tension, and R is the radius of the droplet. For concentrated emulsions, Pal (1998) proposed a modified Mooney model to calculate the relative storage modulus ( $G_r' = G'/G_c'$ ) and loss modulus ( $G_r'' = G''/G_c''$ ),

$$G'_r = exp\left[\frac{K_1\varphi_d}{1 - K_2\varphi_d}\right] \tag{5}$$

$$G_r'' = exp\left[\frac{K_1\varphi_d}{1 - K_2\varphi_d}\right] \tag{6}$$

where  $K_1$  and  $K_2$  are adjustable parameters. Furthermore, Pal (2000) demonstrated that the effect of dispersed phase volume fraction on the viscoelastic characteristics of the oil-water emulsion is due to the interfacial area and the droplet-droplet interaction. However, compared to the Parlierne model, the modified Mooney model will be more accurate to predict the relative storage and loss modulus in all the ranges of the dispersed phase volume fraction.

The viscoelasticity of crude oil and its emulsions were performed on a series of aspects (Li et al.,

2015; Guo et al., 2016). The micro-theory of the changing viscoelastic characteristics was demonstrated through the comparison analysis of crude oil micrographs. The fractions of saturates, aromatics, resins, and asphaltenes are the direct factors on viscoelasticity. The water-in-oil emulsions are emphasized when investigating the viscoelastic characteristics of crude oil and water emulsions. The effect of dispersed phase volume fraction on the viscoelasticity was widely studied (Sharma et al., 2015; Fernandes et al., 2017). It was found that the storage and loss modulus increased as the dispersed volume fraction increased with the exponent rule. In conclusion, most previous works focused only on stable water-in-crude-oil emulsions (Pal, 1999; Vargas et al., 2018). However, the heavy crude oil and water two-phase flow in pipelines always maintain as an unstable dispersed mixture (water-in-oil or oil-in-water) that shows the different viscoelastic characteristics compared to stable emulsions. It will influence the calculating accuracy of the power of the supplying system. So, the unstable characteristics must be considered to improve the theory of viscoelasticity and industrial application. The corresponding research has not been reported in the literature.

In this work, the viscoelastic characteristics of unstable heavy crude-oil-water dispersed mixtures are investigated to improve pipeline transportation. In particular, the unstable characteristic of the oil-water two-phase dispersed mixtures is considered, which has not been investigated in previous work. Two types of heavy crude oils with different compositions are chosen as the research samples to retain the consequence reliability. The small amplitude oscillation measurement method is used to measure the viscoelastic parameters systematically. The effects of temperature, dispersed phase volume fraction, and droplet distribution are considered, as they are important parameters in the study of pipeline transportation systems of heavy crude-oil-water mixtures.

#### 2. Experimental

2.1. Materials

Two types of heavy crude oils are chosen as the research samples to retain the consequence reliability. Crude oil A is produced in the Sui-zhong oilfield, whereas crude oil B is produced in the Bo-hai oilfield in China. They exhibit vastly different physical properties such as density and viscosity. Further, these chosen crude oils are non-Newtonian fluids with different degrees of shear thinning characteristics.

The physical properties of these heavy crude oils are listed in Table 1, and the rules of viscosity vs. temperature are displayed in Fig. 1. The viscosity of these heavy crude oils continually decreases as the temperature increases. It shows that the density and viscosity of crude oil A are bigger than crude oil B. The rheological measurement shows the same results between tap-water-oil and mineral-water-oil dispersed mixtures. Therefore, tap water is used as the water phase; its density is 998 kg/m<sup>3</sup> at 30 °C. The rheological characteristics of crude oil A, such as shear thinning, thixotropy and yield stress, can be found in Zhang et al. (2017).

2.2. Preparation of dispersed mixtures

In this study, a series of stable and unstable oil-water two-phase dispersed mixtures were prepared to consider the effect of dispersed phase distribution and different unstable characteristics. The mixtures were prepared with different oil volume fractions in the range of 0 to 1. A three-blade stirrer was used to homogenize the oil-water two-phase solutions at a fixed speed of 1000 r/min. Further, several different stirring times, including 100 s, 300 s, and 1200 s, were chosen to obtain the series of experimental mixtures with different dispersed phase distributions and unstable characteristics. The separation phenomenon of the prepared unstable crude-oil-water dispersed mixtures was measured. Figure 2 displays the stability of crude oil A and water dispersed mixture, the oil volume fraction is 0.5

and the stirring time is 100 s on preparation. It shows that the prepared mixtures are keeping on stable when the settling time is less than 5 min. And then, the prepared unstable mixtures will begin to separate which can be found in Figure.2(c) and (d) with the settling time of 15 min and 30 min respectively. The oil-water two-phase mixtures are more stable with the increasing stirring time in the present study.

The microstructure of the prepared oil-water two-phase dispersed mixtures was obtained using a trinocular optical microscope Olympas BX43 with an adapted CCD camera ProgRes C5. Subsequently, the dispersed phase distribution was measured through the relevant microstructure pictures. The measurement of the precipitated water from the oil-water dispersed mixtures as the increasing standing time after preparation was used to measure their unstable characteristics quantitatively.

#### 2.3. Experimental methods

The rheological characteristics, including the shear viscosity and viscoelasticity of the oil-water dispersed mixtures, were investigated using the HAAKE RS6000 rheometer that has a coaxial cylinder sensor system, Z38. 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. This rheometer has a range of shear rate from 0.001 to 1500 s<sup>-1</sup>, and a range of viscosity from 0.5 to 10<sup>6</sup> mPa·s. A variety of temperature control units are available to handle experimental temperatures ranging from 0 to 100 °C in the accuracy of 0.1 °C. In this study, the experimental temperature was maintained in the range from 10 to 90 °C, which is typical in the petroleum industry. The rheological measuring time is controlled on less than 3 min, and the time between preparation of dispersion and ending of the rheological measurement is less than 5 min to promise the stability of the prepared unstable oil-water dispersed mixtures during experiment. At the same time, the phase distribution of unstable mixture was

monitored by through the high-speed camera in the process of the similar experiments with a transparent cup which has the same size of cup Z43. The experimental method was displayed in Figure.3, it includes one transparent cup with the 43 mm inner diameter and one transparent water bath which was used to control the measurement temperature and eliminate the light refraction. Therefore, the stability of crude-oil-water dispersed mixtures can be obtained.

The small amplitude oscillatory shear measurement method with the stress control test model (CS) was used to measure the viscoelastic characteristics of heavy crude oil and its dispersed mixtures. In the CS test model, stress is assigned with an amplitude  $\tau_0$  and an angular velocity  $\omega$ , and the corresponding strain is measured with the strain amplitude  $\gamma_0$  and the phase angle  $\delta$ . A shear stress sweep with a constant frequency was first performed to obtain the region of linear viscoelasticity. Subsequently, the frequency sweep was performed to measure the viscoelastic characteristics. At least, three replicates of each test were performed to improve the reliability of the consequences. The error analysis was given on all results.

#### 3. Results and discussion

#### 3.1. Viscoelasticity of heavy crude oil

Heavy crude oil exhibits complex viscoelastic characteristics because of its components of saturates, aromatics, resins, and asphaltenes. The viscoelastic characteristics of crude oils A, and B were measured for the further investigation of their dispersed mixtures. The effect of temperature was also considered.

Fig. 4 displays the shear stress sweep consequence of crude oil B at a constant frequency of 1 Hz. The measurement temperatures are 20 °C, 30 °C, 40 °C, and 70 °C. The measured storage modulus and loss modulus are maintained as the shear stress increases in the measurement condition where the shear

stress is less than 100 Pa. Therefore, crude oil B remains in the linear viscoelasticity region when the shear stress is less than 100 Pa. The experimental temperature slightly affects the region of linear viscoelasticity (Li et al., 2009). The effect of temperature on the value of storage modulus and loss modulus is shown in Fig. 5. The figure shows that the storage modulus of the two types of heavy crude oils is maintained at approximately 0.4 Pa, which does not change as the temperature increases. However, the loss modulus decreases as the temperature increases. Compared to the changing behavior of viscosity, the effect of temperature on the viscosity and loss modulus yields almost the same behavior (Zhang et al., 2013). This is because the parameters of viscosity and loss modulus express the same characteristic, i.e., the shear dissipation of fluids. Further, the two types of heavy crude oils with different components display the same behavior as the temperature changes.

The results of the frequency sweep measurements on the linear viscoelasticity region are displayed in Fig. 6. The chosen shear stress amplitude is 1.0 Pa and maintains in the linear viscoelasticity region. It shows that the storage modulus (G') and loss modulus (G'') increased as the sweep frequency increased continuously. The storage modulus is less than the loss modulus (G'<G'') at the smaller frequency range at first; however, the opposite is shown when the value of the sweep frequency is larger than some points at different temperatures. In other words, the viscoelastic characteristic of heavy crude oil is viscous dominant (G'<G'') in the small sweep frequency, and it changes to elastic dominant (G'>G'') when the sweep frequency increased to a threshold value. Further, the eigenvalue of the complex viscosity increased as the sweep frequency increased.

In this work, the value of frequency when G' = G'' on the sweep frequency measurement is defined as the critical frequency point  $(f_c)$ . The heavy crude oil is viscous dominant (G' < G'') when the sweep frequency is less than the critical frequency point, and changes to the elastic dominant (G' > G'') when

the sweep frequency is larger than the critical point. Fig. 7 shows the effect of temperature on the critical frequency of heavy crude oil A, and B. It shows that the critical frequency continually decreases with the increasing temperature with similar behaviors. In conclusion, a larger system temperature will accelerate the change in the heavy crude oil from viscous dominant to elastic dominant.

3.2. Viscoelasticity of heavy crude-oil-water dispersed mixtures

A series of heavy crude-oil-water dispersed mixtures were prepared with different oil volume fractions and dispersed phase distributions. Different droplet size distributions were obtained by controlling the stirring time. Shear stress sweep and frequency sweep measurements were performed subsequently to measure the linear viscoelastic characteristics of heavy crude-oil-water dispersed mixtures. The effect of oil volume fraction, temperature, and droplet size distribution were considered systematically.

#### 3.2.1. Effect of the phase volume fraction

The stirring parameters of speed and time in preparation of the heavy crude-oil-water dispersed mixtures are 1000 r/min and 300 s, respectively. The oil volume fraction is within the range of 0-1. Further, the result of crude oil B was chosen to be displayed and analyzed.

The microstructure of heavy crude-oil-water dispersed mixtures with the oil volume fractions of 0.2, 0.4, 0.6, and 0.8 is shown in Fig. 8. It shows that water is in the continuous phase (O/W) when the oil volume fractions are 0.2 and 0.4, and oil is in the continuous phase (W/O) when the oil volume fractions are 0.6 and 0.8. Fig. 9 displays the droplet size distributions of the corresponding microstructures. As shown, the droplet size distributions are always shown as a Gaussian distribution. The median diameters  $d_{50}$  of the four oil-water dispersed mixtures with different oil volume fractions are 25 µm, 20 µm, 35 µm, and 35 µm, respectively. The value of the median diameter is larger than the

most of the oil-water stable emulsions (Langevin et al., 2004). Further, the heavy crude-oil-water dispersed mixtures obtained in this study are unstable. Oil and water will separate after stirring which is similar to the pipeline flow in industry.

The results of the frequency sweep measurements of the heavy crude-oil-water dispersed mixtures with different oil volume fractions are displayed in Fig. 10. The chosen shear stress amplitude is 1.0 Pa; therefore, the shear deformation remained in the linear viscoelasticity region. As shown, the oil-water dispersed mixtures exhibit the similar behavior of pure oils; meanwhile, the storage modulus (G') and loss modulus (G'') increased as the sweep frequency increased continuously (Ghannam et al., 2012). The storage modulus is less than the loss modulus (G'<G'') at the smaller frequency range at first; however, the opposite is shown when the value of sweep frequency is larger than some points with different oil volume fractions. In other words, the viscoelastic characteristic of heavy crude-oil-water dispersed mixtures is viscous dominant (G'<G'') in the small sweep frequency, and changes to elastic dominant (G'>G'') when the sweep frequency increased to a threshold value.

The effect of oil volume fraction on the storage modulus (G') and loss modulus (G'') of oil-water dispersed mixtures is shown in Fig. 11. It shows that the storage modulus is maintained as the oil volume fraction changes. The mixing of water into the heavy crude oil slightly affects the storage modulus. The loss modulus increases as the oil volume fraction of the oil-in-water dispersed mixtures increases. And, it will increase to its maximum value when the oil volume fraction increases to the phase inversion point of the oil-water mixtures. Therein, the phase inversion point is defined as the critical oil volume fraction between the water-in-oil and oil-in-water mixtures. The phase inversion point of crude oil B and water mixtures is 0.6 in this study. After the phase inversion point, the loss modulus will decrease as the oil volume fraction increases continuously. As reported in previous

studies, the effect of dispersed phase fraction on the loss modulus can be predicted by the modified Mooney model. In Fig. 11, the predicted value of the loss modulus is displayed as the oil volume fraction varies. The fitting parameters  $K_1$  and  $K_2$  in Eq. 6 are 5.41 and 0.04 for oil-in-water mixtures, and 2.98 and 0.19 for water-in-oil mixtures. It shows that the modified Mooney model can describe the loss modulus data excellently.

As the characteristic of pure heavy crude oil, the oil-water dispersed mixture is viscous dominant (G' < G'') when the sweep frequency is less than the critical frequency point, and changes to elastic dominant (G' > G'') when the sweep frequency is larger than the critical point. Fig. 12 displays the effect of oil volume fraction on the critical frequency. It shows that the critical frequency continually increases as the oil volume fraction for the oil-in-water mixtures increases, and it increases to its peak value on the phase inversion point. Subsequently, the critical frequency decreases as the oil volume fraction increases.

#### 3.2.2. Effect of the system temperature

The microstructure of heavy crude oil and its mixtures is changed owing to different temperatures. Subsequently, the viscoelastic characteristics should be influenced by the changing temperatures that always appear on the heavy crude oil pipeline transportation system. In this section, the effect of temperature on the viscoelasticity of heavy crude-oil-water dispersed mixtures is investigated systematically.

Fig. 13 displays the changing behaviors of the storage modulus, loss modulus, and complex viscosity of heavy crude-oil-water dispersed mixtures with increasing temperature. It shows that the three viscoelastic parameters decrease as the temperature increases. This is because the mechanical strength of the interfacial film decreases when the temperature increases. Further, the ability to resist droplet

deformation owing to shear force becomes weaker as the temperature increases.

To our knowledge, the effect of temperature on viscosity can be described by the Arrhenius model (Eq.

7):

$$\eta = A e^{B/T} \tag{7}$$

where A and B are constant parameters that can be obtained by experimental measurements.

The theory of the effect of temperature on the viscoelasticity and viscosity of oil-water mixtures is the same as through the changing of the microstructure. Therefore, the Arrhenius model was used to describe the changing behaviors of the storage modulus, loss modulus, and complex viscosity as the temperature changes. The predicted consequence is displayed in Fig. 13. It shows that the Arrhenius model can describe the behaviors of the viscoelasticity of heavy crude-oil-water dispersed mixtures with the changing temperature excellently.

The effect of temperature on the critical frequency of heavy crude-oil-water dispersed mixtures was also measured in this study. The experimental consequence is listed in Fig. 14. It shows that the critical frequency continually decreased with increasing temperature, which is similar to pure crude oil. A larger system temperature will accelerate the change in heavy crude-oil-water dispersed mixtures from viscous dominant to elastic dominant.

#### 3.2.3. Effect of the droplet size distribution

The droplet size and its polydispersity in the dispersed phase are important in characterizing the stability of dispersed mixtures, and they are determined by two opposite mechanisms of droplet breakup and coalescence when the mixtures are under the shearing condition. In this work, different stirring times in preparing the mixtures are used to obtain different droplets sizes and polydispersities. The phenomena of droplet coalescence and sedimentation are analyzed to determine the stability of the

experimental oil-water dispersed mixtures. Subsequently, the effect of the droplet size distribution on viscoelasticity of heavy crude-oil-water dispersed mixture is studied in this section.

The microstructure of heavy crude-oil-water dispersed mixtures, obtained by different stirring time, is shown in Fig. 15. The oil volume fraction of these mixtures is 0.5, and the stirring times are 100 s, 300 s, and 1200 s. This shows that these oil-water mixtures remain as water-in-oil emulsions. Fig. 16 displays the droplet size distributions of the corresponding microstructures. It shows that the droplet size distributions are approximately Gaussian. The median diameters  $d_{50}$  of the three oil-water dispersed mixtures obtained by different stirring times are 42 µm, 34.5 µm, and 17 µm. The median diameter decreased as the stirring time increased using the same preparing conditions. The number of droplets increased as the median diameter decreased with the same dispersed phase volume fraction. Therefore, the distance between two droplets will be smaller as the stirring time increases, which can increase the stability of oil-water dispersed mixtures.

The effect of droplet size distribution on the viscoelasticity of heavy crude-oil-water dispersed mixtures has been measured. The changing behaviors of storage modulus G', loss modulus G'', and complex viscosity  $|\eta^*|$  with different median diameters are shown in Fig. 17. As shown in the figure, the viscoelastic parameters increased as the median diameter decreased with the approximate exponent behaviors in this study. The reason is that the link force between two droplets will be increased by the smaller distance through the longer stirring time, which will decrease the median diameter. Further, the smaller droplet, which is more difficult to deform by shear stress, can increase the values of the viscoelastic parameters.

The effect of droplet size distribution on the critical frequency of heavy crude-oil-water dispersed mixtures is listed in Fig. 18. It shows that the critical frequency continually decreases as the median

diameter increases. A larger median diameter will accelerate the change in heavy crude-oil-water dispersed mixtures from viscous dominant to elastic dominant.

## 4. Conclusion

The viscoelastic characteristic of the heavy crude oil and water two-phase dispersion mixture is an important rheological characteristic. In particular, it affects the shutdown and restart sections of pipeline transportation. In this work, the viscoelastic characteristics of unstable heavy crude-oil-water dispersed mixtures were investigated to improve pipeline transportation. The effects of temperature, dispersed phase volume fraction, and droplet size distribution were considered. Additionally, the influence of unstable characteristic on the viscoelasticity was investigated for the first time. It shows that the unstable heavy crude-oil-water dispersed mixture display the great different viscoelastic characteristics compared to the normal crude-oil-water emulsions. The unstable characteristics must be considered in the design of the heavy crude oil and water two-phase dispersed flow system.

The mixing of water into heavy crude oil slightly affected the storage modulus, which maintains a constant value as the oil volume fraction changes. However, the loss modulus increases as the oil volume fraction of the oil-in-water dispersed mixtures increases. Subsequently, the loss modulus decreases with the increasing oil volume fraction after the phase inversion point, which can be described by the modified Mooney model accurately. The viscoelastic parameters decreased as the temperature increased, and the Arrhenius model was suggested to describe the corresponding changing behaviors. The storage and loss modulus were increased as the median diameter decreased with the approximate exponent behaviors. Furthermore, the critical frequency will decrease as the increasing temperature or droplet median diameter. These studies will benefit the pipeline transportation system of heavy crude-oil-water mixtures.

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Oil types	Density (kg/m <sup>3</sup> )	Viscosity (mPas)	SARA analysis (wt.%)			
			Saturates	Aromatics	Resins	Asphaltenes
Crude oil A	955	5439	33.2	26.2	37.2	3.4
Crude oil B	920	242	52.7	27.3	18.4	1.6

Table. 1. Physical properties of experimental oils (T = 30 °C, P = 101.325 kPa,  $\dot{\gamma} = 10$  s<sup>-1</sup>).

Ϋ́



Fig. 1. Viscosity vs. temperature for the experimental heavy crude oils.



Fig.2. Separation phenomenon of the prepared unstable crude-oil-water dispersed mixtures with

different settling time (Crude oil A; oil volume fraction, 0.5; stirring time, 100s).



Fig. 3. The experimental method of stability test of dispersed mixtures in 2.5 mm gap under the shear

condition.



Fig. 4. Shear stress sweep measurements at different temperatures (crude oil B, f = 1 Hz).



Fig. 5. Effect of temperature on storage modulus (G') and loss modulus (G'') on the linear

viscoelasticity region.



Fig. 6. Frequency sweep measurement on the linear viscoelasticity region at different temperature

(crude oil B,  $\tau = 1.0$  Pa).



Fig. 7. Critical frequency point of the elastic and viscous dominated (G' = G'') contained different

temperature ( $\tau = 1.0$  Pa).



Fig. 8. Microstructure of heavy crude-oil-water dispersed mixtures with different oil volume fractions

(crude oil B, stirring time 300 s).



Fig. 9. Droplets size distributions with different oil volume fractions (crude oil B, stirring time 300 s).



Fig. 10. Frequency sweep consequences of oil-water dispersed mixtures with different oil volume

fractions (crude oil B,  $\tau = 1.0$  Pa, T = 30 °C).

![](_page_31_Figure_1.jpeg)

Fig. 11. Effect of oil volume fraction on storage modulus (G') and loss modulus (G'') of oil-water

dispersed mixtures (crude oil B, f = 1 Hz,  $\tau = 1.0$  Pa, T = 30 °C).

![](_page_32_Figure_1.jpeg)

Fig. 12. Critical frequency point of the elastic and viscous dominated (G' = G'') with different oil

volume fractions (crude oil B,  $\tau = 1.0$  Pa, T = 30 °C).

![](_page_33_Figure_1.jpeg)

Fig. 13. Effect of temperature on the viscoelasticity of heavy crude-oil-water dispersed mixtures

(Crude oil A, f = 1 Hz,  $\tau = 1.0$  Pa).

![](_page_34_Figure_1.jpeg)

Fig. 14. Effect of temperature on the critical frequency of heavy crude-oil-water dispersed mixtures

(Crude oil A,  $\tau = 1.0$  Pa).

![](_page_35_Picture_1.jpeg)

Fig. 15. Microstructure of heavy crude-oil-water dispersed mixtures by different stirring times 100 s,

300 s, 1200 s (crude oil A,  $\varepsilon_o = 0.5$ ).

![](_page_36_Figure_1.jpeg)

![](_page_36_Figure_2.jpeg)

times (crude oil A,  $\varepsilon_o = 0.5$ ).

![](_page_37_Figure_1.jpeg)

Fig. 17. Effect of droplet size distribution on the viscoelastic characteristic of heavy crude-oil-water

dispersed mixtures (crude oil A,  $\varepsilon_o = 0.5$ ).

![](_page_38_Figure_1.jpeg)

Fig. 18. Effect of droplet size distribution on the critical frequency of heavy crude-oil-water dispersed

mixtures.

- The viscoelasticity of unstable heavy crude oil-water dispersed mixtures was studied by the microstructure observation and rheological test.
- Storage modulus is always keeping on constant as the changing oil volume fractions.
- Loss modulus increases first, and then decreases as the increasing oil volume fractions. And its peak value appears at the phase inversion point.
- Both storage modulus and loss modulus increased as the decreasing droplet median diameter, and decreased as the increasing temperature.