Rheological Behaviour of Crude Oil: Effect of Temperature and Seawater[1]

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Abstract—The effect of temperature and seawater on rheological behavior was investigated in this work for four samples of crude oil from Algerian Sahara. The Newtonian model was used to fit the shear stress dependence on the shear rate for temperature range between 20 and 100°C. The temperature increase leads to a decrease in the dynamic viscosity of crude oil. The temperature dependence of dynamic viscosity was fitted by the Walther equation. For crude oil emulsion, the Herschel-Bulkley model was used to fit the shear stress dependence on the shear rate for volume fraction of seawater between 30 and 70%. The increase in the volume fraction of seawater induces not only an increase in the yield stress and the consistency index of crude oil emulsions but also a decrease in the flow index of crude oil emulsions.

INTRODUCTION

The knowledge of rheological properties of crude oil and the crude oil-water mixture is important in petroleum industry in order to optimize the process parameters of proper design of pipes and pumping of petroleum crude. For this reason, the understanding of the hydro-dynamical behavior of crude oil and emulsification water in crude oil flows is of the prime importance. According to Zhang et al. [1], the gelation of crude oil causes a disaster during transport of crude oil on long distance in pipeline. Gelled oil is not the common occurrence for pipelines under normal operation. The rheological properties of crude oil and crude oil-water mixtures depend on several factors such as the origin and chemical composition of crude oil [2], pH and temperature [3–5], water content [6] and surfactants [7]. The rheology of crude oil and crude oil in water emulsions has been widely studied and it was the subject of many previous works. The effect of temperature and the presence of surfactants on the rheological properties of Buchan crude oil-water interface have been studied by Mohammed et al. [8]. The authors have shown that interfacial films become more liquid-like at temperatures of 45°C and the increase in the viscosity with increasing temperature. Taborda et al. [9] investigated the effect of nanoparticles and nanofluids as viscosity reducers for heavy crude oil. It was shown that the increasing in the

concentration of nanoparticles in the mixture caused reduction of viscosity approximately by 90% in comparison with the nanoparticle-free crude oil. The authors also demonstrated that the higher concentration of nanoparticles, the effectiveness of the heavy-oil viscosity reduction diminishes. Zhang et al. [10] used the shearing method for measuring the apparent viscosity of crude oil-water mixtures with water fraction. The authors have demonstrated that the degree of dispersing the crude oil-water emulsification depends on shear rate and the oil phase has greater influence on the apparent viscosity of the mixture than the water phase leading to higher viscosity. Therefore, the free water has more effect on the apparent viscosity than oil, leading to a lower viscosity. In order to create of emulsions with an acceptable level of viscosity of crude oil, Malkin et al. [11] studied the morphology and rheological properties of the formed emulsions. The authors have shown the introduction of surfactants with the hydrophilic-lipophilic balance great or equal to fifteen into heavy oil leads to the formation of emulsions with water as a continuous phase. According to these authors the emulsions are viscoplastic and their viscosity in the range of high shear stresses can be sharply reduced in comparison with the viscosity of crude oil. A comparative study of the rheological properties for typical heavy and light crude oils of Russian origin and their mixtures was investigated by Ilyin et al. [12]. The authors have demonstrated that the light oil is a viscoelastic non-Newtonian fluid in presence of paraffin waxes and the presence of crystalliz-

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Table 1. The characteristics of the crude oil samples

Sample	Gravity, °API	Sulfur content, wt %	Density, 15°C	Asphaltenes, wt %	Resins, wt %	Freezing point, °C
1	32.7	0.0792	0.76	0.52	6.44	−33
2	42.6	0.0802	0.78	0.55	6.46	−33
3	47.5	0.0819	0.80	0.58	6.48	−33
4	48.6	0.0829	0.83	0.60	6.50	−33

able waxes in crude oils makes it viscoplastic with yield stress. The authors also shown that above 28°C the light oil becomes a non-elastic Newtonian fluid and heavy oil demonstrates Newtonian behavior even when cooled to −30°C. In contrast, heavy oil becomes viscoelastic at temperatures lower than 0°C.

The main objective of this study is to explore the rheological characteristics of crude oil and seawater crude oil emulsion as a function of temperature and seawater content in order to provide useful information for the proper design of pipes and pumping of petroleum crude.

MATERIALS AND METHODS

Sample Preparation

The crude oil used in this study was obtained from the different regions of Algerian Sahara (Samples 1, 2, 3, and 4) as a liquid. The main characteristics of the crude oil samples are presented in Table 1.

The emulsions used in this study are mixtures of seawater and different crude oil samples, homogenization is obtained by continuous mechanical agitation for 2 h at ambient temperature. Figure 1 shows photomicrographs of emulsion 70% seawater and 30% crude oil.

Experimental Setup

The rheological measurements were performed by using a torque controlled rheometer (RS600 from Thermo-Fischer), equipped with a cone-and-plate geometry (diameter: 60 mm; angle: 2 degree; gap: 105 μm). In order to prevent changes in composition

during measurements due to water evaporation, a solvent trap was placed around the measuring device.

Methods

Effect of temperature on rheological behavior of crude oil. In order to avoid any memory effect, the sample was then kept at rest for 300 s prior to measurements in order to allow the material to recover, at least partially; its initial structure [13]. The flow curves were obtained by applying an increasing shear rate ramp from 0.01 to 200 s^{-1} during 300 s. The experiments were performed at different temperatures between 20 and 100°C. A new fresh sample was used for each temperature in order to avoid any irreversible evolution of the cutting oil emulsion. In order to investigate the reproducibility of results, two replicates were made for most of the experiments. Because the similarity of curves we present a single flow curve for a sample of crude oil.

Effect of seawater on rheological behavior of crude oil. For this study the volume fraction of seawater (30, 50, and 70%) used for each samples of crude oil at 25°C. The flow curves were obtained by applying the increasing shear rate ramp from 0.01 to 200 s^{-1} during 300 s.

RESULTS AND DISCUSSION

Effect of the Temperature on Rheological Behavior of Crude Oil

Effect of temperature on dynamic viscosity of crude oil. The rheological behavior of all samples as a function at different temperatures is Newtonian, the fitted
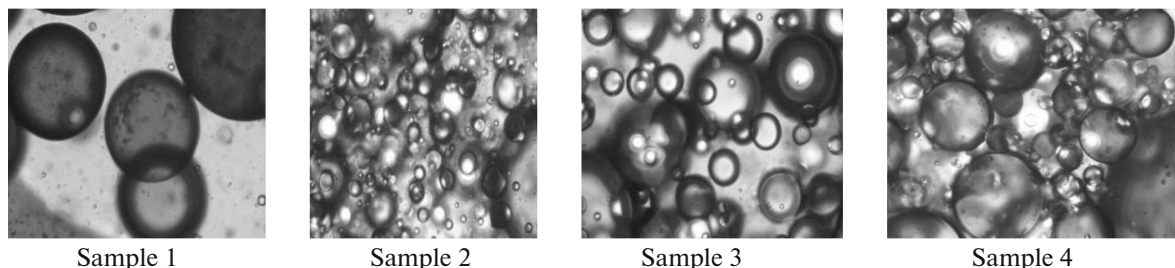

| Sample 1 | Sample 2 | Sample 3 | Sample 4 |

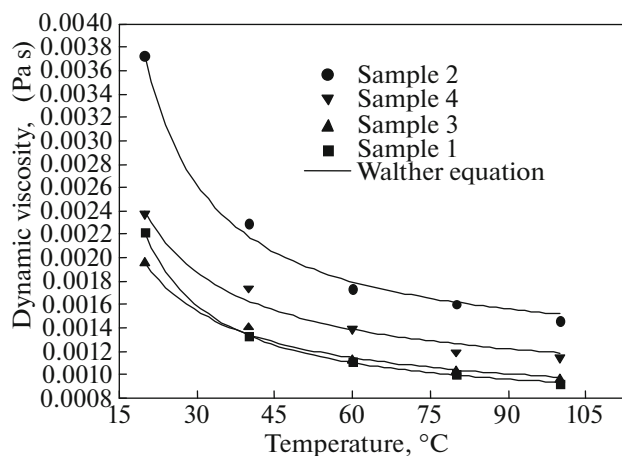**Fig. 1.** Photomicrographs of emulsion 70% seawater and 30% crude oil.

Fig. 2. Dynamic viscosity as a function of temperature for each sample of crude oil. The solid lines correspond to the curve fitting to Eq. (1).

Fig. 3. Shear stress as a function of shear rate at different emulsions seawater sample 4 of crude oil at 25°C. The solid lines correspond to the curve fitting to law Newtonian and Eq. (2).

parameters of the Newtonian law as the function of temperature are presented in Fig. 2. It appears that dynamic viscosity decreases with increasing temperature. Figure 2 shows the dynamic viscosity of the crude oil samples. For all samples of crude oil, a decrease in dynamic viscosity with the increase in temperature is observed from 20 to 100°C. A likely explanation is a weakening of the network strength between particles of the dispersions due to thermal motion. This temperature dependence of the dynamic viscosity can be described by the Walther type equation [14]:

$$\mu = \mu_0 \exp\left[\left(\frac{T_0}{T}\right)^m\right], \quad (1)$$

where T_0: ambient temperature, μ_0: dynamic viscosity at ambient temperature in Pa s and m: a constant that depends on the sample studied.

The fitted parameters of the Walther equation as a function of temperature are presented in Table 2.

Effect of Seawater on Rheological Behavior of Crude Oil

Typical flow curve of the crude oil emulsion. Figure 3 shows an example of the shear stress evolution as a function of shear rate for different emulsions with 0, 30, 50, and 70% of seawater for sample 4 of crude oil. The variation of the shear stress τ as a function of the shear rate $\dot{\gamma}$ for the four studied samples of crude oil clearly shows the Non-Newtonian behavior above a yield stress. Therefore experimental data were fitted to the classical model of Herschel-Bulkley with the coefficients of regression equal to 0.99 for alls samples:

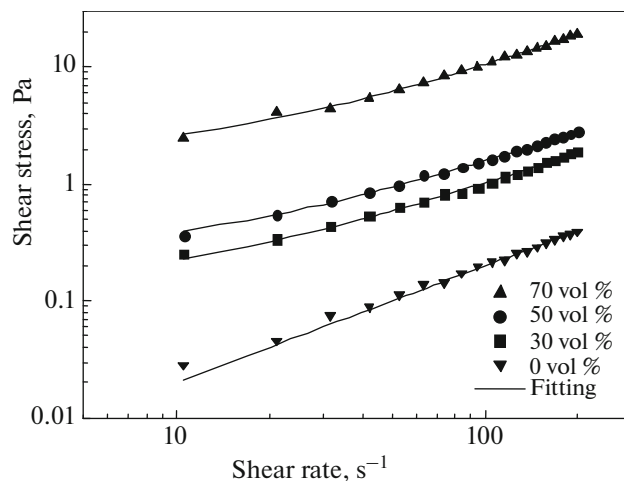$$\tau = \tau_0 + K\dot{\gamma}^n, \quad (2)$$

where τ_0 is the yield stress in Pa, K the consistency index in Pa s^n and n is the flow index.

The fitting parameters of the Herschel-Bukley model as a function of volume fraction of seawater for different samples are presented in Figs. 4 and 5. It appears that the yield stress and consistency index increase with increasing volume fraction of seawater in crude oil. Concerning the flow index, it decreases with increasing volume fraction of seawater in crude oil.

Viscoelastic properties of seawater-crude oil emulsion. The dynamic measurements are an efficient tool for investigating the properties of emulsions of seawater-crude oil under conditions close to at rest state, allowing investigating the viscoelastic properties without rupture of the internal structure of a system. Thus the dynamic measurements could be interpreted in the framework of the linear viscoelastic theory. The elastic moduli $G'(\omega)$ and loss $G''(\omega)$ moduli were measured at frequencies ranging from 0.01 to 100 Hz at the fixed shear stress of 0.2 Pa. Figure 6 shows the evolution of

Table 2. Fitted parameters of the Walther equation of each crude oil sample

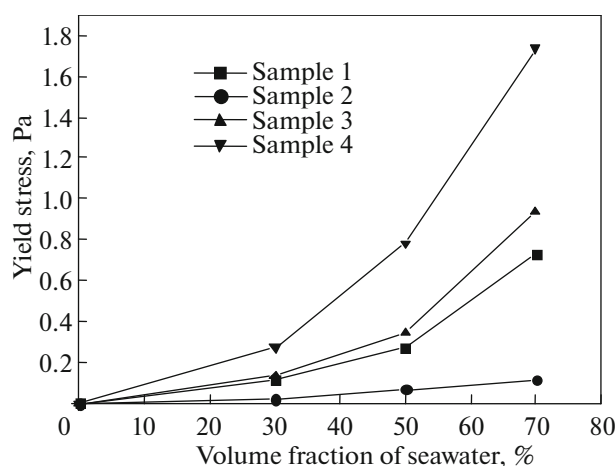| Samples	μ_0, Pa s	$m(-)$	R^2
1	0.00067	0.80	0.99
2	0.00011	0.8505	0.99
3	0.0006	0.5858	0.99
4	0.0007	0.5955	0.98

Fig. 4. Yield stress as a function of seawater added in crude oil.

G' and G'' as a function of frequency for emulsion seawater sample 1 of crude oil.

We observed in Fig. 6 the elastic modulus G' and the loss modulus G'' are strongly frequency dependent. For frequencies below the characteristic frequency [15] which depends on the volume fraction of seawater added to crude oil the elastic moduli G' is less than the loss moduli G'' and the emulsions presented as elastic fluid,when the frequency characteristicis exceeded, elastic moduli G' becomes greater than the loss moduli G'' and emulsion demonstrate viscous fluid behavior. The crossover frequency in which the elastic and loss moduli were equal, has been used to characterize the relative importance of elastic and viscous effects in emulsions in which crossover existed frequency corresponds to the frequency in which the phase angle δ between the imposed stress and resulting strain is 45°; i.e., $\tan(\delta) = \dfrac{G''}{G'} = 1$ [15].

$$t_r = \frac{1}{2\pi f_c}, \qquad (3)$$

where t_r is the relaxation time in s and f_c is the characteristic frequency in Hz.

Table 3 shows the variation of the characteristic frequency and the relaxation time (Eq. (3)) in terms of the volume fraction of seawater add in crude oil. In Table 3 we observe an increase of the characteristic frequency with increasing volume fraction of seawater added to crude oil caused an increase in the viscous domain. This means that the breakdown rate of the flocculation structure in emulsion decreases with increasing volume fraction of seawater add in crude oil. We also observe a rapid decrease of the relaxation time of emulsion with increasing volume fraction of seawater added to crude oil, thus increasing the volume fraction of seawater causes a rapid structured of emulsions. This behavior could be related to the formation of a three-dimensional network. At the highest volume fraction of seawater, the local seawater − crude oil flocculation is the main factor responsible for the structured state of emulsions.

CONCLUSIONS

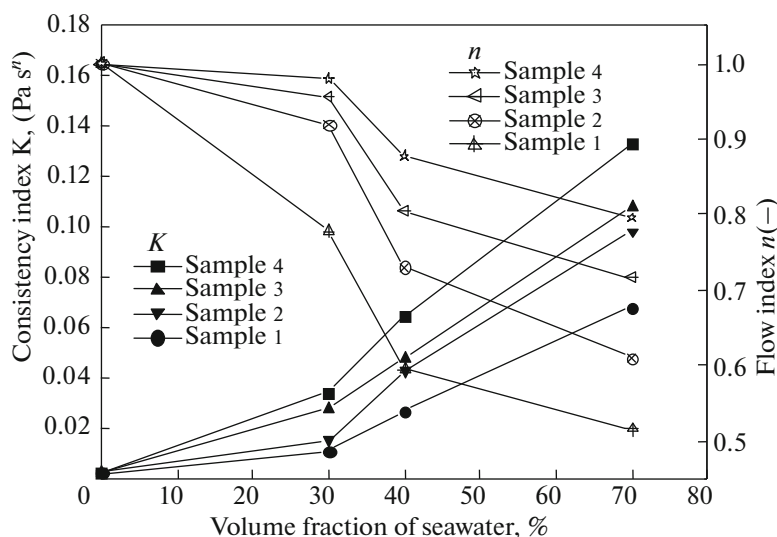In this study, the effect of temperature on the rheological behavior of four samples of crude oil from

Fig. 5. Consistency index and flow index as a function of seawater added in crude oil.
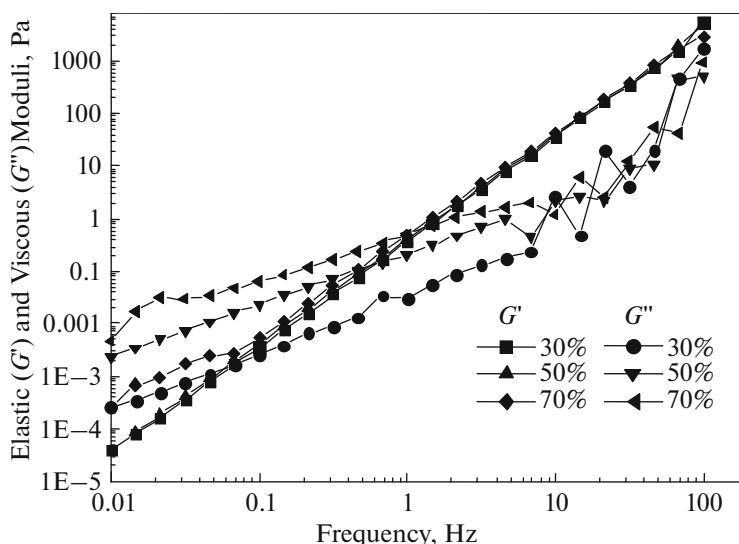
Fig. 6. Variation of elastic (G') and viscous (G'') moduli with oscillation frequency at 25°C and a search stress of 0.2 Pa for seawater sample 1 emulsions.

Algerian Sahara and the effect of seawater added to crude oil have been performed.

The Newtonian model with temperature dependent parameters was found to be well correlated with the behavior of crude oil. The dynamic viscosity decreased with increasing temperature and their relationship could be modeled by the Walther type equation.

The Herschel-Bulkley model with parameters depending on the seawater concentration was found to be well correlated with the behavior of seawater—crude oil emulsions. The yield stress and the consistency index increased with water concentration while the flow index decreased with increase in water concentration.

The addition of seawater into oil causes a change in the rheological behavior of oil: domain viscoelasticity increases with increase in the percentage of seawater in crude oil. A rapid decrease in the relaxation time of emulsion with increasing volume fraction of seawater in crude oil was observed, increasing the volume fraction of seawater causes the rapid structuring of emulsions.

Table 3. Variation of the characteristic frequency and the relaxation time as a function of a volume fraction of seawater add in crude oil

Sample of crude oil	Volume of seawater, %	Characteristic frequency f_c, Hz	Relaxation time t_r, s
	30	0.068	2.335
1	50	0.566	0.281
	70	1.022	0.156
	30	0.322	0.495
2	50	1.318	0.121
	70	1.37	0.116
	30	0.056	2.845
3	50	0.0627	2.540
	70	0.289	0.550

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