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Light crude oil rheology under chemical solvents treatment

Farid SOUAS[*1] and Abderazak GUECIOUER[2]

[1]*LEGHYD Laboratory, Faculty of Civil Engineering, University of Science and Technology Houari Boumediene (USTHB), Bab Ezzouar, Alger, Algeria*
[2]*Research Unit Materials, Processes and Environment (UR-MPE), Faculty of Engineering Science, University M'Hamed Bougara, Boumerdes, Algeria*

Abstract. This work attempts to study the rheological behavior of Algerian light crude oil from Hassi-Messaoud field with and without chemical solvents in order to improve the flow characteristics. Using the rheometer AR2000, an experimental investigation was conducted to measure the rheological properties via flow test and dynamic mode (oscillation) at various temperatures. Several factors such as temperature (20, 30, and 45 °C), shear rate (between 0.01 and 700 s^{-1}), and solvent concentration (between 2 and 6% of toluene, naphtha, and kerosene) on the rheological parameters have all been studied for this purpose. The statistical parameter standard error (SE) provided justification for the experimental validation of the Herschel-Bulkley model. The results of the flow test showed that these solvents had a significant impact on the flow characteristics of light crude oil at various temperatures, with toluene being the most effective. The viscoelastic properties of crude oil were shown to be considerably influenced by temperature and solvent type, as demonstrated by the dynamic mode study that identified the complex modulus (G*), elastic modulus (G′), and viscous modulus (G″).

***Keywords*:** light crude oil; rheology; solvents; temperature; viscosity; viscoelastic behavior.

1. Introduction

Refined fossil fuels are used to make liquefied petroleum gas, diesel, gasoline, and feedstock for petrochemicals. Petroleum products can be used to generate electricity, power for vehicles, and buildings heating, among other things. Petrochemical firms are a subset of the industrial sector that use petroleum to produce various solvents, chemicals, polyurethane, and intermediates. Crude oil will continue to be needed by the chemical industry for the near future since it is too valuable to be used as fuel or for processes like heating, drying, evaporation, distillation, and separation [1]. With its mixture of paraffins, aromatics, asphaltenes, resins, and saturated hydrocarbons, crude oil is a significant fossil fuel. One of the most traded commodities in the modern, expanding globe is crude oil. It significantly influences global politics and the economy [2]. For example, Algeria possesses the ability to produce about 1.4 million barrels of crude oil daily. Algeria is ranked 10th globally, ahead of Ecuador and Qatar, as an average production in OPEC [3]. Under downhole conditions (high temperature and high pressure), the crude oil's flow is Newtonian; but, once it reaches the surface, it starts to behave as a non-Newtonian fluid. The most practical and affordable method of transporting crude oils and products is via pipelines. But flow in pipelines is far more intricate. Heavy substances like wax and asphaltenes accumulate on the pipeline walls as crude oil's temperature drops. Designing pipelines and pump stations both heavily depends on measuring the rheological characteristics of crude oil. Rheological property measurement is essential since crude oil derived from various sources and has varying compositions [4]. In fact, one of the primary factors influencing the safe transportation of crude oil or petroleum oils is their rheological characteristics. Oil businesses plan to avoid engineering issues and optimally leverage current networks in order to achieve this goal. It has been demonstrated that the rheological characteristics have a major impact on the capacity of pipelines to transport crude oil [5]. Many alternative transportation technologies have been employed, such as the creation of core annular flow (CAF) and drag-reducing additives, the dilution or blending of crude with lighter oil or organic solvents, the preheating of crude followed by pipeline heating, the use of pour point depressants (PPDs), and the development of crude oil emulsion in water (O/W). For a particular application, any of these approaches can have logistical, technological, or financial disadvantages [6-10].

The impact of chemical additions on the rheological characteristics of crude oil has been the subject of recent research. In the work by Azeem et al. [11], a synthetic polymeric pour point depressant (PPD) from olive oil (*Olea europaea*) was employed (500 to 1500 ppm concentration) to reduce the pour point and improve the rheology of wax crude oil. After adding 1000 ppm PPD, the pour point, which was initially 43 °C, was reduced by a maximum of 8 °C. Rheology was investigated at various temperatures (35 to 45 °C) and shear rates (0.1–1000 s^{-1}) with or without mixing of synthesized PPD.

[*]Corresponding author. *E-mail addresses:* fa.souas@gmail.com; farid.souas@usthb.edu.dz (Farid Souas)

Viscosity was reduced by 89.20% at 45 °C following a 1000 ppm PPD mixture. It was discovered that the viscoelastic property lowers the cost of heat treatment when transporting crude oil through pipelines in cold climates. Bded and Ahmad [12] examined the effects of a number of variables on crude oil flow, including the shear rates (1.86 - 233 s^{-1}), the temperature (5 - 30 °C), and the different ratios (500, 1000, and 1500 ppm) of the blend of dimethyl ketone (DMK) and methyl esters extracted from vegetable sesame oil (VSO) as flow improver. Results showed that the combination improved low-temperature crude oil flowability. Besides, the viscosity was reduced to 90.5% and 89.8% at 1500 ppm and 1.86 and 46.5 s^{-1} shear rates, respectively. The decrease in viscosity was particularly notable at shear rates exceeding 100 s^{-1} at temperatures up to 30 °C.

The current research focuses on the effect of chemical solvents such as toluene, naphtha, and kerosene on rheological parameters and viscoelastic properties of Algerian light crude oil.

2. Experimental

The Hassi-Messaoud oilfield, situated in southeast Algeria, provided the light crude oil sample. Physicochemical properties such as density, API gravity, kinematic viscosity and SARA analysis were determined as shown in Table 1 (as received).

Table 1. Characteristics of the light crude oil and solvents used in this study.

Parameter	Value
Crude oil, density at 15.6 °C (g/cm^3)	0.800
Crude oil, API gravity	45.2
Kinematic viscosity at 20 °C (cSt)	2.53
Saturates (wt%)	75.57
Aromatics (wt%)	20.36
Resins (wt%)	4.00
Asphaltenes (wt%)	0.07
Naphtha, density at 15.6 °C (g/cm^3)	0.715
Toluene, density at 15.6 °C (g/cm^3)	0.866
Kerosene, density at 15.6 °C (g/cm^3)	0.792

In this work, the Algerian refinery provided the additives toluene, naphtha, and kerosene, with density also shown in Table 1. These solvents were selected based on how well they improved flow properties in previous studies [13-16]. The crude oil was combined with various amounts of these three solvents to create 50 ml samples, which were then studied. The percentage of added solvents ranged from 2 to 6 wt.%. The crude oil was homogenized by shaking it for 15 minutes at 250 rpm in an incubator shaker Heidolph MR 3001k. The sample was heated to a certain temperature for each solvent concentration (various temperatures were applied to fresh samples). Three temperatures: 20, 30, and 45 °C were chosen because this region's crude oil is susceptible to these temperature ranges.

The AR-2000 rheometer (TA- Instrument) with geometry Couette (diameter of 14 mm) was used for all rheological testing, including rheological characteristics and oscillation mode investigation [5, 17-25]. The controlled stress (CS), oscillation (OSC), and universal controlled rate (CR) test modes are the three ways this rheometer can be used. With a gap of 1 mm between the top and lower plates and a 14 mm diameter, cylindrical and Couette geometry were used for the analysis. This kind of instrument has a wide surface area, which allows for good accuracy and the ability to measure very small viscosity values. To ensure proper homogeneity of the samples, each one will undergo a 60-second pre-shear at a shear rate of 0.15 s^{-1} [26]. After giving the samples a minute to settle, the data gathering process begins. The range of the applied shear rate was 0.01 to 700 s^{-1}. The linear viscoelasticity domain was used for the dynamic tests (in oscillation mode) at frequencies between 0.1 and 10 rad/s.

3. Results and discussion

3.1. Rheological model

The light crude oil sample's flow behavior was examined at temperatures ranging from 20 to 45 °C and shear rates between 0.01 and 700 s^{-1}. The measurement was carried out under controlled rate (CR). A typical rheogram in terms of shear stress and shear rate is seen in Figure 1b.

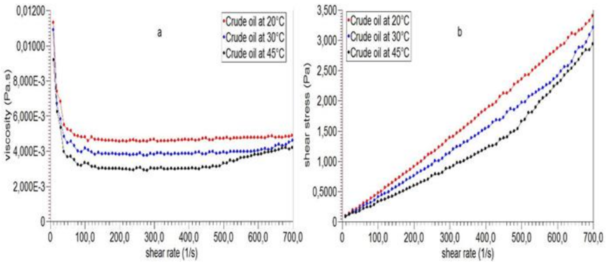

Figure 1. Flow curves of pure crude oil at different temperatures: (a) viscosity (b) shear stress.

With increasing shear rate, the resulting shear stress progressively and considerably increases.

The flow behavior of the crude oil sample was examined using four different rheological models: the power law model, the Herschel and Bulkley model, the Bingham model, and the Casson model, as indicated by Equations 1 - 4.

$$\tau = K\dot{\gamma}^n \quad (1)$$

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

$$\tau = \tau_0 + \mu\dot{\gamma} \quad (3)$$

$$\sqrt{\tau} = \sqrt{\tau_0} + \sqrt{\mu\dot{\gamma}} \quad (4)$$

where: τ is shear stress (Pa); τ_0 is apparent yield stress (Pa); $\dot{\gamma}$ is shear rate (s^{-1}); n is the flow behavior index; K is the consistency index (Pa·s^n) and μ is apparent viscosity (Pa·s).

Resulted equation parameters are given in Table 2.

Table 2. Rheological parameters while dealing with different models.

	t (°C)	τ_0(Pa)	K ($Pa \cdot s^n$)	n	μ ($Pa \cdot s$)	SE
Power law (Eq. 1)	20	-	3.688E-3	1.040	-	8.007
	30	-	1.961E-3	1.117	-	21.58
	45	-	3.392E-4	1.377	-	33.69
Herschel-Bulkley (Eq. 2)	20	0.0970	2.362E-3	1.105	-	**4.173**
	30	0.0920	6.641E-4	1.277	-	**16.59**
	45	0.0834	3.177E-5	1.732	-	**20.48**
Bingham (Eq. 3)	20	2.561E-5	-	-	4.737E-3	10.99
	30	6.881E-5	-	-	4.060E-3	28.75
	45	1.050E-4	-	-	3.578E-3	62.73
Casson (Eq. 4)	20	5.527E-4	-	-	3.362E-5	569,3
	30	5.164E-6	-	-	4.054E-3	28.94
	45	1.323E-3	-	-	3.230E-3	70.01

Between the expected shear stress from the rheological model and the measured shear stress during the experiment, the statistical parameters are estimated in terms of error. The statistical parameter standard error (SE), which can be stated as follows, is computed with Eq. 5 in order to evaluate the correctness of rheological models.

$$SE = \left[\frac{\left[\frac{\sum_{i=1}^{n}(x_m - x_c)^2}{n-2}\right]^{\frac{1}{2}}}{X_m^{max} - X_m^{min}} \right] \times 1000 \qquad (5)$$

where: x_m is the measured value; x_c is the calculated value and n is the number of data points.

Based on the modeling study results presented in Table 2, and taking into account the minimal standard errors, it can be concluded that the Herschel Bulkley model provides the most accurate description of crude flow behavior throughout a known range of shear rates. The behavior index (n) has values greater than 1, and it is also evident that the index (n) rises as temperature rises. It can be inferred that crude oil has pseudoplastic behavior at various temperatures [27, 28].

3.2. Effect of temperature on viscosity

The viscosity and other viscous properties of light crude oil are strongly influenced by temperature. The study examined the impact of temperature on viscosity within the 20 – 45 °C temperature range, as depicted in Figure 1 (a). It is evident that the crude oil exhibits a non-Newtonian pseudoplastic nature at specific temperatures, as seen in Figure 1 b. In fact, when the applied shear stress is less than a certain value (referred to as yield stress), no flow occurs. When temperatures rise from 20 to 45 °C, there are noticeable differences in the viscosity of crude oil. Additionally, it has been seen in experiments that when temperature rises, yield stress (τ) and shear stress both decrease.

The term "extent of viscosity reduction" (EVR%) was established to express the drop in viscosity. The EVR% is determined as follows:

$$EVR\% = \frac{(N_r - N_c)}{N_r} \times 100 \qquad (6)$$

where: N_r is reference viscosity (viscosity value at which all viscosity measurements were obtained for comparison) at 288.5 s^{-1} shear rate and 20 °C; N_c is the viscosity at 288.5 s^{-1} shear rate and corresponding temperature. The EVR% for the 20–45 °C temperature range is shown in Table 3.

Table 3. Viscosity and EVR% of crude oil versus temperature at a shear rate of 288.5 s^{-1} with different concentrations of solvents.

	Temperature					
	20 °C		30 °C		45 °C	
	Viscosity (mPa·s)	EVR (%)	Viscosity (mPa·s)	EVR (%)	Viscosity (mPa·s)	EVR (%)
Pure crude	4.64	0	3.85	**17.02**	3.04	**34.42**
Crude + 2% Kerosene	3.97	**14.45**	3.33	**28.20**	2.58	**44.44**
Crude + 4% Kerosene	4.07	**12.26**	3.38	**27.10**	2.63	**43.28**
Crude + 6% Kerosene	4.08	**12.13**	3.33	**28.20**	2.65	**42.91**
Crude + 2% Naphta	3.95	**14.82**	3.30	**28.95**	2.56	**44.79**
Crude + 4% Naphta	3.93	**15.30**	3.34	**28.11**	2.55	**45.02**
Crude + 6% Naphta	3.94	**15.08**	3.23	**30.42**	2.53	**45.54**
Crude + 2% Toluene	3.89	**16.20**	3.29	**29.06**	2.66	**42.70**
Crude + 4% Toluene	3.88	**16.24**	3.28	**29.23**	2.62	**43.62**
Crude + 6% Toluene	3.46	**25.46**	3.04	**34.55**	2.33	**49.89**

The EVR% increased noticeably to 34.4% during the studied temperature range. There could be a number of contributing elements behind this. First of all, the viscosity of the heavy components in crude oil is greatly influenced by temperature. Viscosity decreases as a result of high temperatures destroying the heavy components' predefined structure in the crude oil phase [29–31]. It was also discovered that the shear rate affects the viscosity of light crude oil. At high shear rates, the viscosity drops to low values. This implies that with larger shear rates, there is less resistance to the flow. This is because light crude oil contains molecular chains. Light crude oil viscosity decreases as the shear rate rises because the chain type molecule becomes disentangled and reorients parallel to the driving force [2, 16, 32].

3.3. Effect of the addition of various solvents on the viscosity of light crude oil

Figures 2-10 show the flow behavior of the system light crude oil/ solvents that has been studied and plotted.

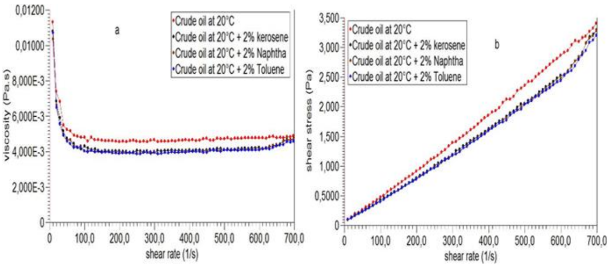

Figure 2. Flow curves of crude oil treated with 2% solvents at 20 °C.

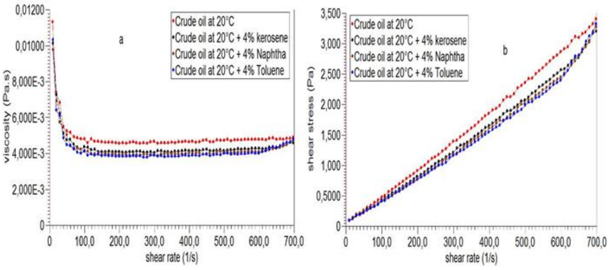

Figure 3. Flow curves of crude oil treated with 4% solvents at 20 °C.

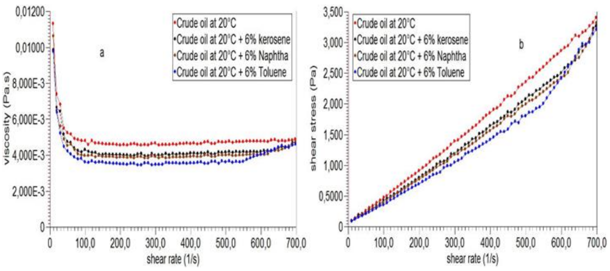

Figure 4. Flow curves of crude oil treated with 6% solvents at 20 °C.

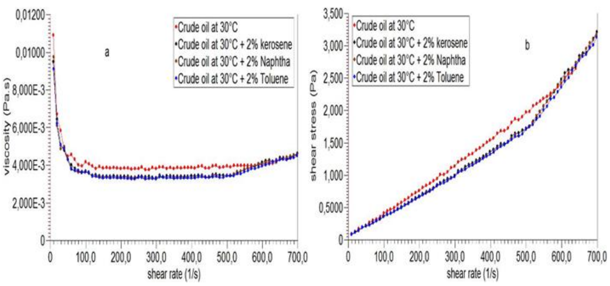

Figure 5. Flow curves of crude oil treated with 2% solvents at 30 °C.

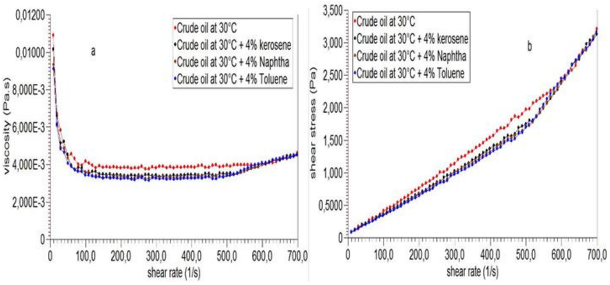

Figure 6. Flow curves of crude oil treated with 4% solvents at 30 °C.

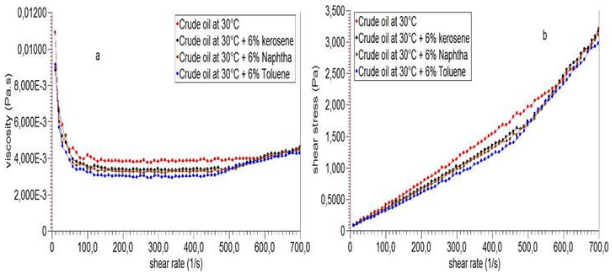

Figure 7. Flow curves of crude oil treated with 6% solvents at 30 °C.

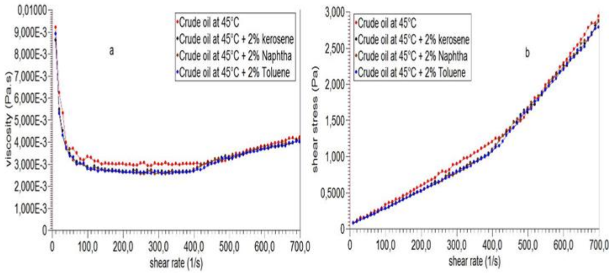

Figure 8. Flow curves of crude oil treated with 2% solvents at 45 °C.

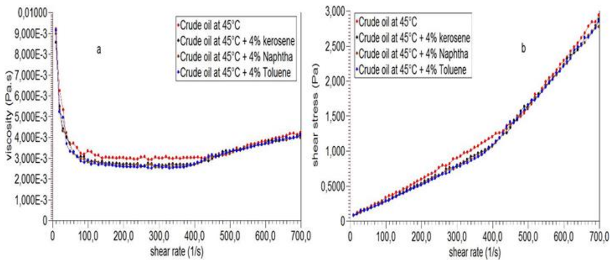

Figure 9. Flow curves of crude oil treated with 4% solvents at 45 °C.

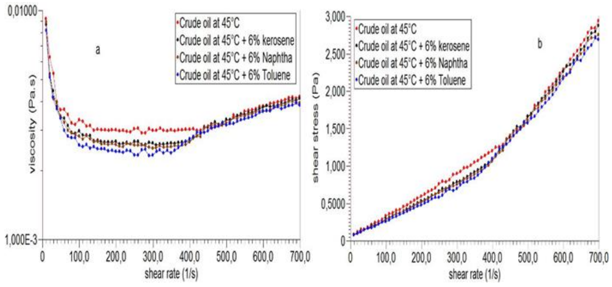

Figure 10. Flow curves of crude oil treated with 6% solvents at 45 °C.

The obtained results demonstrate that the apparent viscosity and shear stress of the samples at the three examined temperatures can greatly decrease by adding only a small quantity of each of the three solvents, but they still converge to an asymptotic value at higher shear rates. This decrease is influenced by the solvent type, test temperature, and applied shear rate in addition to the solvent concentration. It should be mentioned that when the concentration of solvents in the solution increases, the rate at which the viscosity reduces may somewhat drop. The solvent solution's saturation provides an explanation for this.

As indicated in Table 3, the viscosity of light crude drops by 14.45% and 14.82%, respectively, when 2% kerosene and naphtha are added at 20 °C. At the same temperature and concentration, toluene causes a 16.20% decrease in viscosity. By raising the concentration to 6%, the extent of viscosity reduction (EVR%) was observed to be 12.13%, 15.08%, and 25.46%, respectively. This indicates that even at lower temperatures, there is a noticeable reduction in viscosity. In each case, the EVR% rises noticeably as the temperature reaches 30 °C. After adding 2% of kerosene and naphtha, respectively, the EVR% rises to 44.44% and 44.79% when the temperature is raised from 20 °C to 45 °C, while it increases to 42.70% for toluene.

At 45 °C, a higher degree of viscosity reduction was seen, which can be attributed to the combined effect of solvents and temperature. During the studies, it was also discovered that the decrease of viscosity at 20 °C with the addition of 6% toluene was even more than when the crude oil was heated to 30 °C for pure crude oil, and at 30 °C it was even equal to when the crude oil was heated to 45 °C. When it comes to reducing the viscosity of crude oil, toluene works better than naphtha and kerosene. Under the same conditions, it was found that kerosene was less effective at reducing viscosity. It is evident that these solvents have a significant impact on the rheology of light crude oil, which is highly advantageous for pipeline transportation of light crude oil in situations where pressure drop is a significant problem.

3.4. Viscoelastic studies

A valuable method for comprehending a sample's rheology is the viscoelastic analysis. It includes the impact of oscillating strain or stress on a sample of crude oil. The contribution of stress energy momentarily stored in the sample during the test and that is recoverable is represented by the elastic modulus, or G′. The viscous modulus, or G″, is a measure of the irreversible energy needed to start a flow. When the value of G′ is higher than G″, it indicates that the sample primarily behaves as a solid because of the creation of a gel structure. Complex modulus (G*), which indicates the sample's resistance to applied strain, is an additional factor.

Figure 11 shows the complex modulus vs frequency for light crude oil at various temperatures.

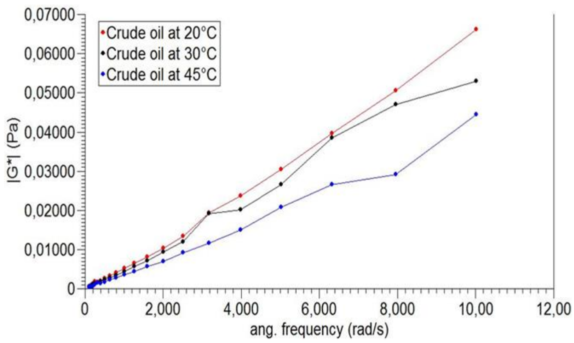

Figure 11. Complex modulus of light crude oil at different temperatures

At every temperature, the measured G* steadily rises with frequency. The entire resistance of the substance against the applied strain reduces dramatically with temperature, as indicated by the large reduction in the complex modulus with temperature [30].

The frequency dependency profiles of G′ and G″ moduli for light crude oil at 20 °C are displayed in Figure 12. The progressive rises in G′ and G″ with frequency are explained by the light crude oil's linear relationship response over the investigated frequency range. The measurement findings demonstrate that the values of the storage modulus, G′, are surpassed by the values of the loss modulus, G″. This indicates that less energy is stored in light crude oil than is lost as heat during a cycle. As a result, light crude oil has a tendency to act more like a viscous liquid than a solid (gel) [30, 33, 34].

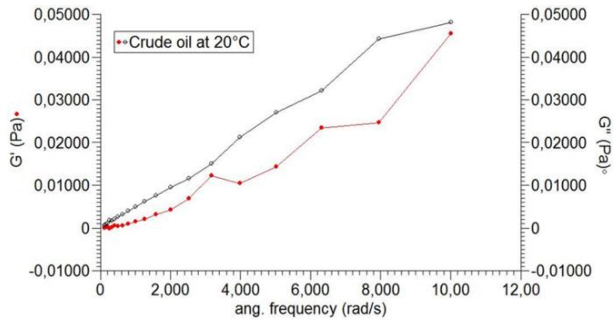

Figure 12. Elastic and viscous moduli of pure crude oil at a temperature of 20 °C

The frequency dependency plots of G′ and G″ for the mixture of pure crude oil and crude oil with 2% solvents at room temperature (20 °C) are shown in Figure 13.

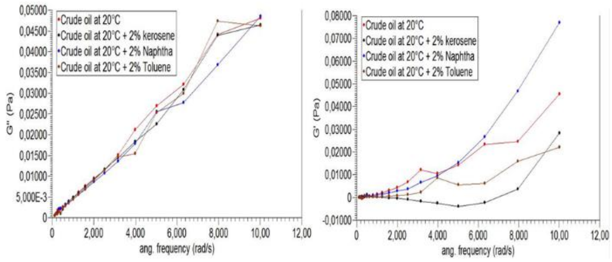

Figure 13. Elastic and viscous moduli of crude oil treated with 2% solvents at 20°C.

The viscous modulus is barely affected by the addition of 2% solvents, but the elastic modulus is significantly altered. However, it should be noted that the frequency has a greater impact on these results. Both toluene and kerosene are effective at lowering elastic modulus G′, and this indicates that more energy is lost as heat at a 2% concentration of each solvent. However, storage modulus in the presence of 2% naphtha yields a greater value than that for pure crude oil with frequency above the value of 5.011 rad/s. The greatest outcomes have been seen with 2% kerosene because of a more significant decrease in G′, which prevents the creation of a gel-like structure.

4. Conclusions

This study aimed to compare the effects of kerosene, naphtha, and toluene on rheological parameters, including viscosity, shear stress, complex modulus, elastic modulus, and viscous modulus. The investigation leads to the following conclusions.

The examined crude oil exhibits rheological behavior in accordance with the Herschel–Bulkley rheological model, which exhibits a yield stress for the flow. Light crude oil encounters a considerable drop in viscosity and shear stress throughout the temperature range of 20, 30 and 45 °C, as well as in shear rate between 0.01 and 700 s^{-1}. The addition of various amounts of three solvents had a substantial effect on the rheological properties of the investigated light crude oil. The flow test revealed that toluene was the most effective at improving flow characteristics, while kerosene was less effective. The dynamic mode showed that the temperature has a considerable effect on the light crude oil's complex modulus. By adding certain amounts of three solvents, the light crude oil improves its elastic character and acts more like a viscous liquid than a solid (gel). Compared to the other two solvents, kerosene is the most effective at enhancing the viscoelastic characteristics of the studied crude oil.

Conflict of interest

The authors declare that there is no conflict of interest regarding the publication of this article.

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