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RHEOLOGICAL CHARACTERIZATION OF ALGERIAN CRUDE OILS

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ABSTRACT. This study examines the rheological behavior of three Algerian crude oils, focusing on steady flow, yield stress, and thixotropic behavior across temperatures. The oils exhibit non-Newtonian pseudoplastic behavior with a yield stress at the onset of flow. Results show that viscosity and shear stress decrease with temperature, reducing yield stress and improving flow. The Herschel-Bulkley model best fits crude oil 1, while the Casson model is more suitable for oils 2 and 3 at 30°C, and the power law and Herschel-Bulkley models apply at 20°C. Over the 20–50°C range, apparent viscosity and shear stress decrease by 71%, 77%, and 79% for oils 1, 2, and 3, respectively, while yield stress drops by 24.4%, 48.5%, and 54%. Thixotropic behavior is also observed, with reduced hysteresis area at higher temperatures, indicating reduced internal friction. These findings highlight the role of temperature in enhancing crude oil flow properties, suggesting heat treatment improves transportability.

Keywords: *Crude oil, rheology, temperature, thixotropy, viscosity, yield stress.*

INTRODUCTION

Energy acquisition remains a critical objective for nations worldwide, with crude oil representing 37% of the global energy supply, positioning it as the primary energy source. Historically, hydrocarbons have served as the

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dominant commercial energy source, continuing through the late 20th century. Crude oil is a complex mixture of various hydrocarbons, which gives rise to a diverse set of physical and chemical properties depending on its composition. These characteristics vary based on the oil's specific content, including asphaltenes, saturated hydrocarbons, aromatics, and resins. Crude oils can be classified into four categories—light, medium, heavy, and extra heavy—based on their properties, including sulfur content, specific gravity (API), and density [1,2]. Specifically, crude oil from Algeria is classified as light based on average values of these parameters [3].

Pipeline transportation is the most practical and cost-effective method for moving crude oil and its derivatives. However, crude oil transport via pipelines is complicated by the inherent variability in its physical and chemical properties, which can change in response to climatic conditions. These variations affect the flow characteristics and transport behavior of crude oil. The non-Newtonian nature of crude oil, attributed to its complex chemical composition and the presence of suspended particles, further complicates long-distance pumping operations. These particles tend to deposit as fouling layers in various facilities, including heat exchangers, fired heaters, oil reservoir rocks, and transfer lines, leading to reduced energy recovery and increased operational costs [4]. The rheological properties of petroleum oils are crucial in several processes involving fluid movement, including migration within reservoirs and transportation through pipelines. Understanding the viscosity and flow behavior of crude oil is essential for designing and optimizing the infrastructure necessary for production and refining operations [5]. Therefore, accurate measurement of the rheological properties of crude oil is vital for the development of pumping stations and pipeline systems, as these properties can vary significantly depending on the crude oil's origin.

The rheological behavior of crude oil is determined by the relationship between shear stress and shear rate, typically characterized by experimental rheometry. A linear relationship indicates Newtonian fluid behavior, while a nonlinear relationship signifies non-Newtonian behavior. Several models have been suggested to describe the rheological behavior of crude oils, including the Herschel-Bulkley, power law, Bingham, and Casson models [6]. Barskaya et al. [7] demonstrate in their study that, despite two crude oils having identical concentrations of resins, asphaltenes, and solid paraffins, they exhibit distinct rheological behaviors at low shear rates. The chemical composition of the high molecular weight components, namely asphaltenes and solid paraffins, was analyzed and revealed no significant differences in their composition. The study identifies that the differing rheological behaviors are attributed to variations in the content of saturated and aromatic hydrocarbons. An increase in mono- and bi-aromatic hydrocarbon content leads to an enhanced non-

Newtonian behavior, which is attributed to the formation of coagulated structures within the crude oil. The authors conclude that these findings should be considered when predicting crude oil viscosity for the development of oil recovery and transportation technologies. Ilyin and Strelets [8] investigated the rheological properties of eight different types of crude oils, including light, heavy, waxy, and extra-heavy oils, using a RheoStress 600 rotational rheometer. Their study demonstrated that the viscosity of crude oils is influenced by their composition, temperature, and the applied shear stress. The authors concluded that the rheological behavior of crude oils is primarily determined by their inherent characteristics.

The purpose of this article is to provide a comprehensive analysis of the rheological behavior of Algerian crude oils, focusing on key aspects such as steady-state rheology, yield stress, and thixotropy. Through a series of experimental investigations, the relationship between viscosity, shear stress, and shear rate at various temperatures is explored, offering valuable insights into the flow characteristics of these crude oils under different conditions. Furthermore, the impact of temperature on viscosity and shear stress at variable shear rates is examined to better understand the thermal sensitivity of these fluids. The article aims to shed light on the yield stress behavior of Algerian crude oils, highlighting their ability to resist flow under low-shear conditions. Additionally, the study addresses the thixotropic properties of these oils, which are essential for understanding their behavior during processing and transportation. By providing a detailed exploration of these key rheological parameters, the article contributes to a deeper understanding of the flow properties of Algerian crude oils, with implications for both the oil industry and the development of improved handling and processing techniques.

RESULTS AND DISCUSSION

Viscosity and shear stress vs. shear-rate at different temperatures.

The flow behavior of all crude oil samples was analyzed using the CR mode of the AR-2000 rheometer, focusing on viscosity versus shear rate and shear stress versus shear rate. The results of the rheological tests are presented in Figures 1 and 2, which illustrate the variation of viscosity and shear stress with shear rate over a temperature range of 20°C and 30°C.

The flow curves demonstrate the non-Newtonian pseudoplastic behavior of the crude oil samples across the entire temperature range. Specifically, the curves indicate a lack of flow when the applied shear stress is below a critical threshold, referred to as the yield point. Moreover, the flow curves exhibit consistent trends across all test temperatures, characterized by a gradual increase in shear stress as the shear rate rises. Experimental observations also reveal that as temperature increases, both viscosity and shear stress decrease, with a corresponding reduction in yield stress.

The experimental data presented in Figures 1-2a demonstrate that the apparent viscosity of the crude oil samples exhibits shear rate dependence.

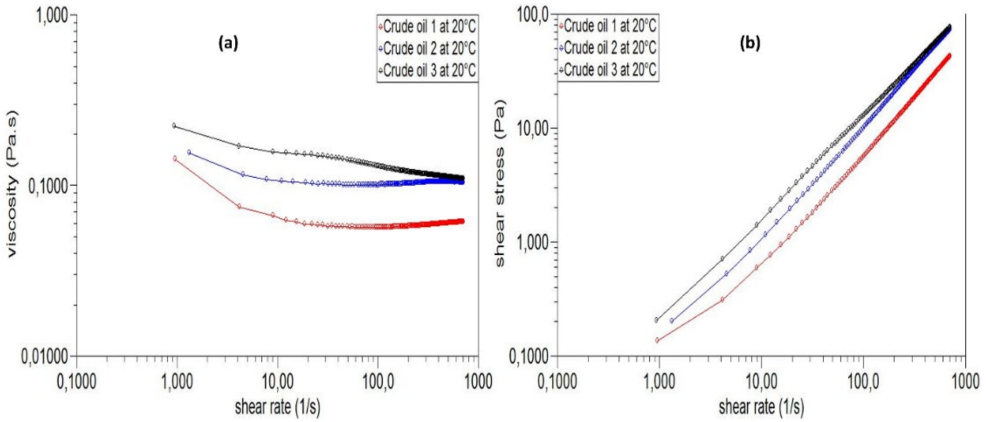

Figure 1. Flow curves of crude oils at a temperature of 20 °C: (a) viscosity vs. shear rate, (b) shear stress vs. shear rate.

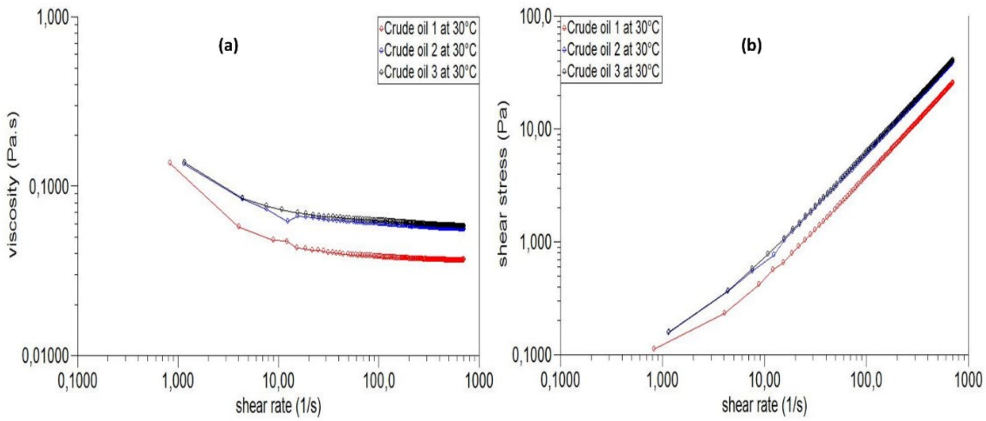

Figure 2. Flow curves of crude oils at a temperature of 30 °C: (a) viscosity vs. shear rate, (b) shear stress vs. shear rate.

The behavior can be categorized into two distinct regions: the first region, observed at low shear rates, is characterized by a significant decrease in viscosity, while the second region, observed at high shear rates, shows a stabilization of apparent viscosity, remaining constant as the heavier components undergo irreversible dissipation at elevated shear rates. Additionally, the viscosity is found to be more sensitive to temperature variations at high shear rates. Notably, a pronounced viscosity reduction is observed between 20°C and 30°C within the test temperature range. The results further indicate that the viscosity decreases more rapidly at lower shear rates and temperatures compared to higher shear rates and temperatures. This effect is particularly evident in crude oil sample 3, which exhibits slightly higher density and heavier components relative to the other samples. These findings are consistent with the results reported by several researchers [9-11] for different crude oil types.

The composition of crude oil samples plays a crucial role in understanding their rheological behavior. Temperature is a key factor influencing the viscosity of heavier constituents in crude oils, such as asphaltenes and resins, with significant effects typically observed within the temperature range of 20 to 30°C. The reduction in viscosity at higher temperatures can be attributed to the thermal influence on the chemical structure of these heavy components. Specifically, the elevated temperature disrupts the ordered structures of the heavy constituents, leading to a substantial and irreversible reduction in viscosity [12-13]. Additionally, the molecular chains present in crude oil samples contribute to viscosity changes. As the shear rate increases, the viscosity of crude oils decreases due to the detangling, stretching, and reorientation of chain-like molecules, which align parallel to the applied force [14].

A modeling analysis was conducted to identify an appropriate rheological model that best fits the experimental measurements. Four distinct rheological models were evaluated in this study, as described by equations 1-4: the Bingham model, the Casson model, the power law model, and the Herschel-Bulkley model.

$$\tau = \tau_0 + \mu\dot{\gamma} \tag{1}$$

$$\sqrt{\tau} = \sqrt{\tau_0} + \sqrt{\mu\dot{\gamma}} \tag{2}$$

$$\tau = K\dot{\gamma}^n \tag{3}$$

$$\tau = \tau_0 + K\dot{\gamma}^n \tag{4}$$

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

The rheological models previously outlined were employed to fit the shear stress data corresponding to various shear rates for each crude oil sample analyzed in this study. The predicted shear stress values from the rheological models and the experimentally measured shear stress are utilized to estimate statistical parameters in terms of error. To assess the accuracy of the rheological models, the standard error (SE), a statistical metric, is calculated as follows [15]:

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

The term x_m represents the measured value, x_c denotes the calculated value, and n refers to the total number of data points.

Table 1. Standard error values of shear stress for various rheological models.

Crude oil	Temperature, °C	Bingham (Eq.1)	Casson (Eq.2)	Power law (Eq.3)	Herschel-Bulkley (Eq.4)
1	20	8.637	10.86	1.418	**0.892**
	30	1.374	1.396	1.969	**1.272**
2	20	3.319	4.056	**3.103**	3.229
	30	2.654	**0.9615**	1.514	1.202
3	20	6.745	2.117	1.638	**1.464**
	30	2.615	**0.9282**	1.485	1.182

Based on the results from the modeling study presented in Table 1, and considering the minimal standard errors, it can be concluded that the Herschel-Bulkley model best describes the flow behavior of crude oil 1 across a known range of shear rates and temperatures. For a temperature of 30°C, the Casson model accurately describes the similar flow behavior of crude oils 2 and 3. However, at 20°C, the power law and Herschel-Bulkley models provide the best fits for crude oils 2 and 3, respectively.

Viscosity and shear stress vs. temperature with variable shear rate.

The relationship between apparent viscosity and shear stress of crude oils as a function of temperature at various shear rates, in order to observe their non-Newtonian behavior, is presented in Table 2. Pressure drop refers to the loss of flow energy caused by the friction between the fluid and the internal pipe walls. Lower viscosity leads to a reduced pressure drop, whereas higher viscosity results in a significant pressure drop and greater dissipation of fluid flow energy. Shear stress is strongly influenced by the viscosity of the crude oil and serves as a measure of the resistance to flow near the pipe walls, where frictional forces occur [16].

Table 2. Influence of temperature on the rheological properties of crude oils at varying shear rates.

		Temperature, °C					
		20°C		30°C		50°C	
Type	Shear rate, S^{-1}	Viscosity, mPa.s	Shear stress, Pa	Viscosity, mPa.s	Shear stress, Pa	Viscosity, mPa.s	Shear stress, Pa
Crude oil 1	100	56.92	5.61	38.39	3.778	17.74	1.754
	200	57.68	11.41	37.37	7.386	17.33	3.433
	300	58.68	17.52	36.9	11.01	17.21	5.143
	400	59.45	23.65	36.65	14.57	17.17	6.835
	500	60.26	30.04	36.55	18.22	17.13	8.547
	600	60.73	36.3	36.52	21.83	17.23	10.31
	700	61.18	42.73	36.62	25.58	17.31	12.1
Crude oil 2	100	99.34	9.826	60.18	5.943	24.5	2.415
	200	102.8	20.37	58.27	11.53	23.82	4.711
	300	104.5	31.24	57.15	17.07	23.63	7.054
	400	105.1	41.84	56.48	22.48	23.49	9.344
	500	105	52.22	56.14	28	23.43	11.68
	600	104.7	62.61	55.98	33.47	23.52	14.06
	700	103.9	72.6	55.84	39.02	23.56	16.46
Crude oil 3	100	129.7	12.77	62.61	6.182	25.59	2.518
	200	120	23.73	60.62	12	24.9	4,921
	300	116.3	34.71	59.45	17.76	24.6	7.342
	400	113.8	45.25	58.77	23.39	24.55	9.759
	500	111.9	55.81	58.52	29.19	24.57	12.25
	600	110.5	66.07	58.22	34.81	24.52	14.66
	700	109.1	76.21	58.1	40.6	24.57	17.16

The crude oils exhibited the greatest sensitivity to heat treatment in the temperature range of 20°C to 50°C. Within this range, a sharp decrease in both apparent viscosity and shear stress was observed, indicating significant changes in the flow properties.

To evaluate the enhancement in the flow behavior of crude oils, specifically in terms of viscosity and shear stress reduction, the Average Degree of Reduction (DAR) is defined. This can be determined using the following equation:

$$(\mathrm{DAR})\% = \frac{1}{n}\sum_{i=1}^{n}\left[\frac{\text{initial value} - \text{final value}}{\text{initial value}}\right] \times 100 \qquad (6)$$

Table 3. Percentage reduction in viscosity and shear stress of crude oils with increasing temperature.

		Temperature, °C					
		20°C - 30°C		30°C - 50°C		20°C - 50°C	
Type	Shear rate, S^{-1}	% Viscosity reduction	% Shear stress reduction	% Viscosity reduction	% Shear stress reduction	% Viscosity reduction	% Shear stress reduction
Crude oil 1	100	32.55	32.66	53.79	53.57	68.83	68.73
	200	35.21	35.27	53.63	53.52	69.95	69.91
	300	37.12	37.16	53.36	53.29	70.67	70.64
	400	38.35	38.40	53.15	53.09	71.12	71.10
	500	39.35	39.35	53.13	53.09	71.57	71.55
	600	39.86	39.86	52.82	52.77	71.63	71.60
	700	40.14	40.14	52.73	52.70	71.71	71.68
Crude oil 2	100	39.42	39.52	59.29	59.36	75.34	75.42
	200	43.32	43.40	59.12	59.14	76.83	76.87
	300	45.31	45.36	58.65	58.68	77.39	77.42
	400	46.26	46.27	58.41	58.43	77.65	77.67
	500	46.53	46.38	58.27	58.29	77.69	77.63
	600	46.53	46.54	57.98	57.99	77.54	77.54
	700	46.26	46.25	57.81	57.82	77.32	77.33
Crude oil 3	100	51.73	51.59	59.13	59.27	80.27	80.28
	200	49.48	49.43	58.92	58.99	79.25	79.26
	300	48.88	48.83	58.62	58.66	78.85	78.85
	400	48.36	48.31	58.23	58.28	78.43	78.43
	500	47.70	47.70	58.01	58.03	78.04	78.05
	600	47.31	47.31	57.88	57.89	77.81	77.81
	700	46.75	46.73	57.71	57.73	77.48	77.48

The results presented in Table 3 show that increasing the temperature from 20°C to 50°C resulted in an average reduction in the initial viscosity and shear stress between the flowing fluid and the pipe wall for crude oils 1, 2, and 3 by approximately 71%, 77%, and 79%, respectively, thereby enhancing transportability. When the temperature was increased from 20°C to 30°C, the reductions were 38%, 45%, and 49%, respectively. From 30°C to 50°C, the reductions were 53%, 59%, and 58%, respectively. The reduction in viscosity is primarily attributed to the temperature-induced decrease in the viscosity of higher molecular weight components, such as wax and asphaltenes, which contributes to a lower viscosity of the entire crude oil mixture. Additionally, the elevation in temperature enhances the Brownian motion of particles, further disrupting the ordered structures of these higher molecular weight components, thereby facilitating a reduction in overall viscosity.

Yield stress

The yield stress is defined as the critical stress threshold beyond which a material transitions from solid-like behavior to fluid-like behavior. Below this threshold, the material exhibits elastic deformation that is directly proportional to the applied stress, following a linear relationship. When the applied stress exceeds the yield stress, the material undergoes irreversible deformation, leading to the initiation of flow [3, 17].

Yield stress measurements of crude oil samples were performed at various temperatures using an AR-2000 Rheometer. This experimental approach enables precise determination of the yield stress by graphically or numerically extrapolating the shear stress versus shear rate (flow curves) to zero shear rate.

Table 4. Yield stress and hysteresis area measurements of crude oils as a function of temperature.

Crude oil	Temperature, °C	Yield stress, Pa	Hysteresis area, Pa/s
Crude oil 1	20	0.135	1375
	30	0.112	212
	50	0.102	117.4
Crude oil 2	20	0.202	2504
	30	0.155	334
	50	0.104	155
Crude oil 3	20	0.210	2975
	30	0.160	345.4
	50	0.097	154.4

Table 4 illustrates the variation in yield stress as a function of temperature for the crude oils. It is evident that the yield stress, which is necessary to initiate flow, decreases with an increase in temperature. For crude oil 1, the yield stress decreased by 24.4% (from 0.135 Pa to 0.102 Pa) as the temperature increased from 20°C to 50°C. Similarly, in the same temperature range, the yield stress for crude oil 2 and crude oil 3 decreased by 48.5% (from 0.202 Pa to 0.104 Pa) and 54% (from 0.210 Pa to 0.1 Pa), respectively. These observations suggest a significant reduction in apparent yield stress with increasing temperature, indicating a decrease in the energy required for the crude oils to flow. This behavior implies that higher temperatures reduce the internal friction, facilitating the flow of the crude oil samples.

Thixotropy behavior

Solutions exhibiting microstructure typically demonstrate thixotropic behavior, characterized by reversible changes between different microstructural states. These changes are governed by the interplay between flow-induced breakdown, buildup from nonflow collisions, and Brownian motion. These mechanisms collectively influence the viscosity of the solution, with each process requiring a certain amount of time to transition between states under applied shear. The thixotropic behavior is commonly quantified by measuring the hysteresis loop, which represents the area between the ascending and descending curves of the rheogram. This area, referred to as the thixotropic area, serves as a key parameter for evaluating thixotropy [18,19,5]. To fully dismantle the thixotropic structure of the crude oil samples, multiple cycles of up and down shear may be necessary to determine the appropriate duration for each cycle. Experimental tests were conducted on the crude oil samples using cycle durations of 100, 200, and 300 seconds for both the up and down shear phases. The shear rate was gradually increased from 0 to 700 s^{-1} over the specified duration and then immediately decreased from 700 to 0 s^{-1} over an equal time period. The hysteresis area was calculated for each cycle. The test utilizing a 300-second duration for both the up and down shear cycles (from 0 to 700 s^{-1} and from 700 to 0 s^{-1}, respectively) was sufficient to ensure complete breakdown of the crude oil structure.

The objective of this study is to examine the thixotropic behavior of crude oil samples. Thixotropy measurements were performed using the CR-mode, with the shear rate incrementally increased from 0 to 700 s^{-1}. The shear stresses and corresponding shear rates were recorded to construct the up-curve of the flow curve. To generate the down-curve, the shear rate was progressively reduced from 700 to 0 s^{-1}. Each cycle had a duration of 300 seconds. For non-thixotropic materials, the down-curve closely mirrors the up-curve. However, for thixotropic materials, the down-curve deviates from the up-curve, creating a hysteresis loop. The area of this loop, denoted as A (in Pa/s), serves as an indicator of thixotropy, providing a measure of the energy per unit volume required to disrupt the thixotropic structure.

Figure 3 illustrates the thixotropic behavior of the crude oils at various temperatures, highlighting the hysteresis phenomenon. The region between the upward and downward curves quantifies the thixotropic effect. This area was measured for each crude oil at the different temperatures tested and is reported as a function of temperature in Table 4. The data, presented in both the figure and table, indicate a significant reduction in thixotropic behavior, with values decreasing from 1375, 2504, and 2975 Pa/s to 117.4, 155, and 154.40 Pa/s for crude oils 1, 2, and 3, respectively, across the temperature range of 20–50 °C. The results indicate that increased temperature reduces the energy required to break bonds, thereby enhancing flow within the pipeline.

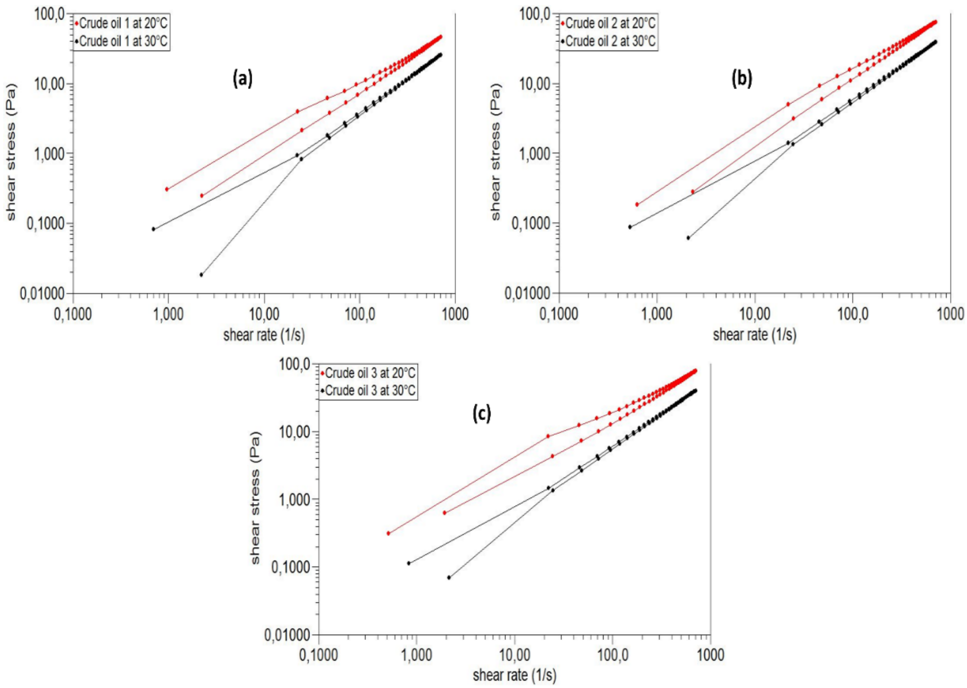

Figure 3. Thixotropic behavior of crude oils at temperatures of 20°C and 30°C.

When a thixotropic structural material is introduced into a viscometer, its microstructure initially consists of large flocs when at rest. As the shear rate is gradually increased, and sufficient time is allowed, the floc size continuously diminishes. At high shear rates, the flocs disintegrate into their individual components. This disaggregation process occurs over a specific period. Conversely, if the shear rate is suddenly reduced, the individual particles begin to colloid and flocculate, progressively forming larger flocs until they reach a size consistent with the new shear conditions. This reassembly process also unfolds over time but at a rate distinct from the breakdown process [18,5].

CONCLUSIONS

In conclusion, the rheological analysis of the three Algerian crude oil samples revealed consistent non-Newtonian pseudoplastic behavior across all tested temperatures, with significant influence from temperature and shear rate on the oils' flow properties. As temperature increased, both the viscosity and yield stress decreased, facilitating easier flow and reducing the internal friction within the crude oils. The apparent viscosity of the crude oils demonstrated shear rate dependence, with the highest sensitivity to temperature observed in the 20°C to 30°C range. Additionally, the study highlighted the distinct rheological behaviors of the samples, with crude oil 3 exhibiting a stronger response due to its heavier components.

The modeling results indicated that different models best describe the flow behavior of the crude oils under various conditions, with the Herschel-Bulkley model accurately capturing the flow of crude oil 1, while the Casson and power law models were most appropriate for crude oils 2 and 3 under certain temperature conditions. Notably, temperature had a pronounced effect on viscosity reduction, with the most significant decreases observed between 20°C and 50°C, improving the transportability of the crude oils. The yield stress also decreased significantly with temperature, further reducing the energy required for flow initiation.

Moreover, the crude oils exhibited thixotropic behavior, which was notably reduced at higher temperatures, suggesting that increased temperatures not only lowered viscosity but also decreased the energy needed to break internal bonds, thus enhancing the flow within pipelines. These findings are crucial for understanding the flow behavior of Algerian crude oils under varying temperature conditions and optimizing their transportation and processing.

EXPERIMENTAL SECTION

Crude oil samples were obtained from multiple oil fields situated in the Tin Fouye Tabankort/South Algeria region (TFT). These samples were labeled as 1, 2, and 3 for identification purposes. The key properties of the crude oil samples are presented in Table 5. The flow behavior of these samples was evaluated using an AR-2000 rheometer [20-23] equipped with a Couette geometry (14 mm diameter) at different temperatures (20, 30 and 50°C). Data acquisition and analysis were conducted using software from TA Instruments (Rheology Advantage Data Analysis Program). The rheometer utilized cylindrical and Couette geometries with a 14 mm diameter and a 1 mm gap between the upper and lower plates. This configuration offers a large surface area, ensuring precise measurements even at very low viscosities. Prior to testing, all samples underwent pre-shear for 30 seconds at a shear rate of 100 s^{-1} to ensure proper homogenization [24]. The samples were allowed to rest for one minute before initiating the measurement process. Shear rates ranging from 0 to 700 s^{-1} were applied during the tests.

Table 5. Properties of the crude oil samples utilized in this study.

Sample	Density, 15°C	Gravity, °API	Asphaltenes, wt %	Resins, wt %
1	0.830	39	0.58	6.50
2	0.840	37	0.62	7.10
3	0.850	35	0.70	7.50

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