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Rheological behavior of oil sludge from Algerian refinery storage tanks

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ABSTRACT

The consumption and demand for petroleum are increasing dramatically with the rapid development of industry and energy sector. As a result, petroleum refineries produce the greatest amount of oily sludge formed at the bottom of storage tanks during oil storage operations, which has a severely negative impact on the storage capacity and the operational safety of the storage tank. The present study focuses on the rheology of this complex fluid from Algerian crude oil storage tanks. Rheological measurements were performed at different temperatures under steady shear and dynamic oscillometry using AR-2000 Rheometer. The results obtained show that the sludge exhibits yield-pseudoplastic flow behavior at low shear rates, which is adequately described by the Herschel Bulkley model based on the standard error and correlation coefficient values. However, quasi-Newtonian flow behavior occurs at very high shear rates. The increase in temperature had positive effects on the rheological properties of the sludge, including dynamic viscosity, shear stress, yield stress, complex modulus, elastic modulus and viscous modulus. The dynamic rheology studies have shown that the sludge material behaves more like a solid than a liquid under all experimental conditions studied.

1. Introduction

Most of the crude oils that are transported to the refinery tend to separate into the heavier and lighter hydrocarbons that make up the crude oil. This problem is frequently exacerbated by cool temperatures, the removal of volatile components and the static state of the crude during storage. The heavy fractions that separate from the crude oil and settle at the bottom of the storage tanks are called "sludge". Sludge is a combination of oil, sediment, heavy metals paraffin, and water. It usually exists in a complex form of water-in-oil emulsion (i.e. water droplets dispersed in oil). Sludge can accelerate corrosion, reduce storage capacity, block tank discharge lines, and disrupt operations, making it necessary to remove these deposits periodically (Heath et al., 2004). However, the sludge should be considered a valuable compound due to its high percentage of oil fractions, as it can be recycled in refineries to reuse the oil recovered from sludge to improve energy resources (Hassanzadeh et al., 2018; Taiwo and Otolorin, 2016). Depending on the petroleum sludge source, its composition can be quite varied, but based on the most reported concentrations, it consists of 4–7% solid sediment and has a higher aliphatic content (40–60%) than aromatic content (25–40%) (Ramirez Guerrero, 2017). Due to the high oil content, various crude oil recovery technologies have been developed for oil sludge treatment, such as solvent extraction, centrifugation, surfactant enhanced oil recovery (EOR), freeze/thaw, pyrolysis, microwave irradiation, electrokinetic, ultrasonic irradiation and froth flotation, etc. (Hu et al., 2013; Hui et al., 2020). The high stability of oily sludge is due to the adsorption of oil on solid particles, resulting in the formation of a protective layer. The presence of polar fractions in the oil, such as resins and asphaltenes, enhances this stability and increases sludge viscosity (Lima et al., 2011). Paraffin can also form a paraffin-based crude oil sludge. This type of sludge is formed during the flocculation of heavier straight-chain hydrocarbons (heavier than C_{20}). They accumulate at the bottom of the reservoir in the form of a viscous gel whose concentration will increase by evaporation of the volatile components, resulting in increased density, viscosity and reduced mobility (Hassanzadeh et al., 2018). The recovery and recycling of oil sludge in the petroleum industry for the optimization of energy resources are mainly conditioned by the chemical composition and viscosity of the sludge. In the tank bottom sludge treatment process, stable material transport is a prerequisite for the normal

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operation of the entire system. Since the tank bottom sludge is usually semi-solid and has poor mobility, it is necessary to study its flow characteristics to guide the design of the pipeline transportation system (Jie et al., 2015). Therefore, the rheological properties of oil sludge are very useful in all processes of its transfer from one site to another (Ghannam et al., 2012).

Rheology is the measurement of the deformation of a system under shear conditions. When the shear rate varies, the system generally exhibits one of four major rheological profiles: Newtonian, dilatant, pseudoplastic and thixotropic. The viscosity of the system with a Newtonian rheological profile will not change when the shear rate is applied or released. In a non-Newtonian group, pseudo-plastic fluids, which are sometimes called shear thinning fluids, play an important role in the oil industry. The viscosity of a pseudoplastic material is high at low shear rates, low at high shear rates, and recovers immediately after the shear rate is released. The dependence of steady-state rheology on temperature and shear rate can be best observed by dynamic rheology tests, which can confirm the development of the microstructural network in crude oil components (Maghzi et al., 2013; Anto et al., 2020). Steady shear rheology, i.e., viscosity and shear stress versus shear rate at different temperatures or viscosity and shear stress versus temperature with variable shear rate, determines the ability of a material to resist structural rupture upon the application of shear energy, while dynamic rheology in terms of response to different angular frequencies allows characterization of existing microstructures without disturbing them in the process, as measurements are limited to small deformations in the linear viscoelastic region (LVR) of the material (Ilyin et al., 2016; Ilyin and Strelets, 2018; Souas et al., 2020, 2021). These measurements are most often successfully performed using rheometers by continuous ramp and frequency sweep tests with a well-defined test protocol.

Abivin et al. (2012) performed detailed steady-state shear experiments and dynamic oscillatory tests of a group of 13 heavy oils of different origins over a range of temperatures. They found that regardless of their zero-shear viscosity, heavy oils have different rheological properties, ranging from a Newtonian and purely viscous character to a weak gel-like behavior related to an elastic internal structure. The study also revealed that for some oils, the viscoelastic character is linked to the presence of the amount of paraffin wax crystals and for the other viscoelastic oils, the elastic character seems to be related to their high amount of asphaltenes.

Unlike heavy crude oil, the solid particles and water content in the sludge at the bottom of the tank are relatively high, which greatly affects the rheological properties of sludge. Therefore, a detailed rheological study of oil sludge is necessary to better understand its behavior and contribute to its management and treatment processes. Although rheology is an essential experimental tool for identifying the interactions between different molecules and characterizing the flow behavior of such a complex material, there are few studies on the flow characteristics and viscosity reduction of oil sludge from tank bottom and on the rheological properties of the oil extracted from the sludge. Hassanzadeh al. (2018) stated that oil sludge from the bottom of the reservoir resulting from the precipitation of heavy molecules of crude oil can cause many problems with significant economic loss. By investigating the effective viscosity reduction factors, a pumpable sludge emulsion can be prepared. Thus, the cleaning of oil storage tanks can be easily done. Therefore, they studied different chemical and physical factors affecting the prepared emulsions (composed of sludge, water and surfactant), such as surfactants, solvents, temperature, pressure and mixing conditions. They found that nonionic surfactants (such as bitumen emulsifier) and solvents (such as mixed xylene, AW-400 and AW-402), applying pressure and mixing operations had a positive effect on reducing emulsion viscosity. Jie et al. (2015) conducted an experimental study on the flow behavior of oil sludge from tank bottom and different methods of reducing its viscosity using HAKKE VT550 rotational viscometer. Several factors such as temperature (20–60 °C), shear rate (0-600s^{-1}), addition of surfactant Triton X-100 aqueous solution, and addition of organic solvents of 1-pentanol and 120# solvent oil on flowability enhancement were studied. The results indicated that within the range of shear rate examined, the oil sludge exhibited the characteristics of both plastic and pseudoplastic fluid. The significant shear-thinning behavior can be attributed to the high solid particles content in the oil sludge. The modeling analysis showed that the Casson model fits the oil sludge flow behavior curve well with the highest coefficient of determination. From the comparison, this study showed that mixing the sludge with a limited amount of organic solvent (120# solvent oil) is the most effective method. Mansur et al. (2015) presented in the rheology section the measurement of rheological properties of crude oil extract from tank bottom sludge. HR3 (Hybrid Discovery) rheometer with Parallel plate (40 mm smart swap, stainless steel) geometry was used to measure the rheological properties of the sample extract under flow test (temperature range of 35–50 °C and shear rate between 10 and 500 s^{-1}) and dynamic test (constant temperature of 50 °C and angular frequency of 1–100 rad/s). They concluded that crude oil exhibits non-Newtonian behavior (shear thinning) over the range of shear rate examined, in which viscosity decreases significantly with increasing temperature due to the effect of temperature on the chemical structure of crude oil. Dynamic rheology data showed that the storage modulus and loss modulus are angular frequency dependent. The extracted oil behaved like a solid rather than a viscous liquid, which could be due to the large solid particles or solid waste present in the oil.

The purpose of the present experimental work is to study the rheological properties of oil sludge from the storage tanks of an Algerian refinery. Indeed, a better understanding of the oil sludge rheology is important for the performance of sludge handling in the petroleum industry, including the economics of the process. This study is divided into three detailed parts. The first is the steady shear experiment as reported in most of the scientific literature on rheological data of materials. The second part of the current study consists of verifying different rheological models such as power law, Herschel and Bulkley, Bingham and Casson to simulate the flow behavior and thus determine the yield stress measurements of the sludge at different temperatures. The third part includes the dynamic rheology of the sludge (viscoelastic behavior).

2. Experimental

2.1. Materials used and sample preparation

The oily sludge (Fig. 1b) used in the experiments was collected from the bottom of the crude oil tank in the oil refinery located east of Algiers, which receives crude oil from different parts of the Algerian desert. After collection, the sticky liquid sludge was stored in a sealed glass jar at room temperature. It was stirred well manually before being used in the experiments and further homogenized by shaking in a Heidolph MR 3001 k model incubator shaker at 250 rpm at 20 °C for 15 min to prepare a 20 ml sample. Table 1 lists some characteristics of the oil sludge used.

2.2. Test methods and protocol

2.2.1. Rheological measurements

All rheological tests (dynamic and continuous flow) were performed using the AR-2000 (Djemiat et al., 2015; Souas et al., 2018a, 2018b, 2019) Couette geometry rheometer with associated

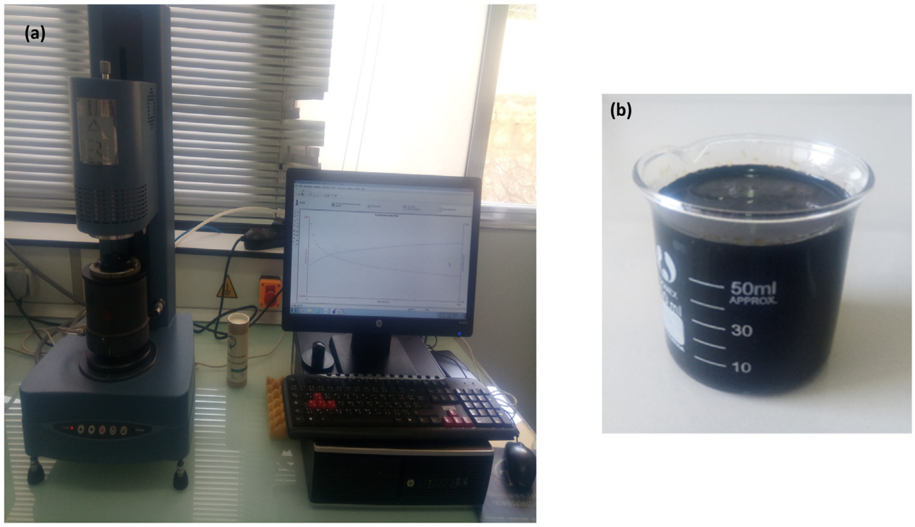

Fig. 1. (a) AR-2000 Rheometer. (b) Oily sludge sample.

Table 1
Characterization of selected sludge from the Algiers refinery.

Characteristics	Amount	Experimental method
oil (wt %)	75	The oil content of the sludge was extracted by the solvent extraction method (Toluene).
water (wt %)	19	The water content was measured by the ASTM D-95 method.
solid particles (wt %)	6	The solid content was calculated by difference.
Metal element (mg/kg)		
V	0.8	The metal composition is determined by Spectrometric analysis according to ASTM D6595.
Fe	300	
Pb	94	
Elemental content (wt %)		
C	77	The elemental analysis is performed in the Thermo Finnigan Flash Model 1112, EA-CHNS/O equipment.
H	12.10	
O	8.20	
N	–	
S	–	
SARA analysis (wt %)		
Saturates	52.10	SARA analysis was performed according to ASTM D2007, by gas chromatography (HP 6890 capillary with FID flame ionization detector).
Aromatics	25.90	
Resins	15.50	
Asphaltenes	6.50	

computer software (Rheology Advantage Data Analysis Program, TA Instruments) (Fig. 1a). A water bath was used to control the temperatures applied in the AR-2000 system. To prevent water evaporation, a cover was placed around the Couette geometry. This rheometer has several test modes of operation: a universal controlled rate mode (CR), a controlled stress mode (CS) and an oscillation test mode (OSC). Due to its large surface area, good accuracy can be achieved with this type of instrument and measurements can be made for very small viscosity values. In addition, at the start of each test, several measurements were made to verify the reproducibility of the AR-2000 rheometer system and the results were very satisfactory.

2.2.2. Testing protocol

Rheological measurements were made under steady shear and oscillatory tests to assess both flow and dynamic properties. In the steady shear test, flow curves in terms of shear stress and viscosity versus shear rate from 0.01 to 700 s^{-1} were recorded at different temperatures using the test protocol previously described in detail by (Djemiat et al., 2015). For the oscillatory test, all dynamic frequency behavior of sludge sample was recorded in the linearity or linear viscoelastic region (LVR) which indicates that the internal bonds of the sample structure are still intact or in other words, shear thinning has not occurred, resulting in the irreversible loss of much of the energy introduced as heat. Subsequently, dynamic mode experiments were performed using a frequency sweep test in the range of 0.1–70 rad/s at different temperatures (30 °C, 40 °C and 50 °C).

3. Results and discussion

3.1. Steady-state rheology

Over time, the properties of the stored crude oil can be altered due to the continuous unloading of the reservoir, pressure variations, changes in temperature and weather conditions, the presence of oxidizing bacteria and fungi, etc. In addition, the light components can evaporate and cause changes in overall composition of the crude oil, the polarity and solubility of the components and their density, the ratio of saturated to aromatic hydrocarbons. As a result, heavy components such as paraffins, asphaltenes, resins and inorganic solids settle at the bottom of storage tanks (Hassanzadeh et al., 2018). The flow behavior of viscous sludge from the bottom of storage tanks will be altered due to structural changes in asphaltene and wax crystals that exhibit nested gel-like structures at lower temperatures. Knowledge of the effect of different parameters on rheological behavior is necessary for analysis of flow modeling, study of complex behavior, pipeline design, flow treatment equipment, reduction of energy consumption, ensuring safety, profitability and processing goals in the petroleum industry.

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

A plot of the apparent viscosity and shear stress of the sludge as a function of temperature at different shear rates to observe the non-Newtonian character was made and shown in Fig. 2a and b. Pressure drop is the loss of flow energy due to the friction of the fluid with the internal walls of the pipe. Lower viscosity results in lower pressure drop, while higher viscosity results in huge pressure drop and dissipation of fluid flow energy. The shear stress is highly dependent on the viscosity of the oil and represents the resistance to flow near the pipe walls where friction occurs (Ibrahim et al., 2017). The sludge showed maximum response in the heat treatment range of 10 °C–50 °C. In this temperature range, there was an abrupt drop in apparent viscosity and shear stress, which indicates alterations in the flow properties.

To assess the improvement in sludge flow characterization in terms of reduction in viscosity and shear stress, the average degree of reduction (DAR) is introduced that can be calculated using the following equation:

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

It was observed that increasing the temperature from 10 to 50 °C leads to an average reduction in the initial sludge viscosity and shear stress between the flowing fluid and the pipe wall of about 59%, which ultimately improves the transportability. While when increasing the temperature from 10 to 30 °C, 46% of the average reduction was achieved and this reduction was halved when increasing the temperature from 30 °C to 50 °C. The increase in temperature reduces the viscosity of the higher weight components of the sludge sample such as wax and asphaltenes to promote a reduction in the viscosity of the whole mixed system, which usually occurs at a temperature of 20–30 °C (Ghannam and Esmail, 2006). Another reason is the intensification of Brownian motion of particles in the system by heating, which further destroys the ordered structure of higher molecular weight components (Jie et al., 2015; Khan, 1996).

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

The variation of viscosity and shear stress of the sludge sample as a function of shear rate (i.e. the flow curves) for the selected temperatures (30 °C, 40 °C, and 50 °C) are presented in Fig. 3. The plot of shear stress versus shear rate (Fig. 3a) indicates the presence of a definite yield point below which flow cannot occur, and then a linear relationship between shear stress and shear rate is observed. Indeed, the fluid needs a finite shear stress to exceed the yield point and initiate flow, suggesting that the sludge sample is non-Newtonian pseudoplastic in nature. It is observed experimentally from the graphs that the shear stress, viscosity and yield stress of the sludge sample decrease with increasing temperature due to the weakening of intermolecular forces with continuous heating (Sharma et al., 2019).

The experimental results in Fig. 3b show the presence of two distinct regions: the low shear rate region up to 300 s^{-1} where the sludge viscosity exhibits strong non-Newtonian shear thinning behavior, and the high shear rate region >300 s^{-1} where the sludge viscosity shows a Newtonian profile. For the second high shear rate region, all curves converge to form a master curve, meaning that the viscosity becomes independent of shear rate and temperature (Hasan et al., 2010). According to Jie et al. (2015), compared to the rheological properties of heavy oil, the shear thinning behavior of sludge is more evident, which is mainly determined by its composition in the tank bottom. As can be seen in Table 1, in addition to the high content of heavy components (asphaltenes and resins), the sludge also contains a significant percentage of solid particles, the presence of which affects the rheological properties of the whole system. The more particles there are, the more pronounced the shear thinning phenomenon is. In the W/O mixture system, particles-particles and particles-oil are tightly intertwined, forming a continuous mesh structure that fills the entire space. This structure together with the aggregate structure of the higher molecular weight components contribute to the high viscosity of the sludge.

When the sludge starts to flow under shear stress, these complex structures are gradually disintegrated and the degree of

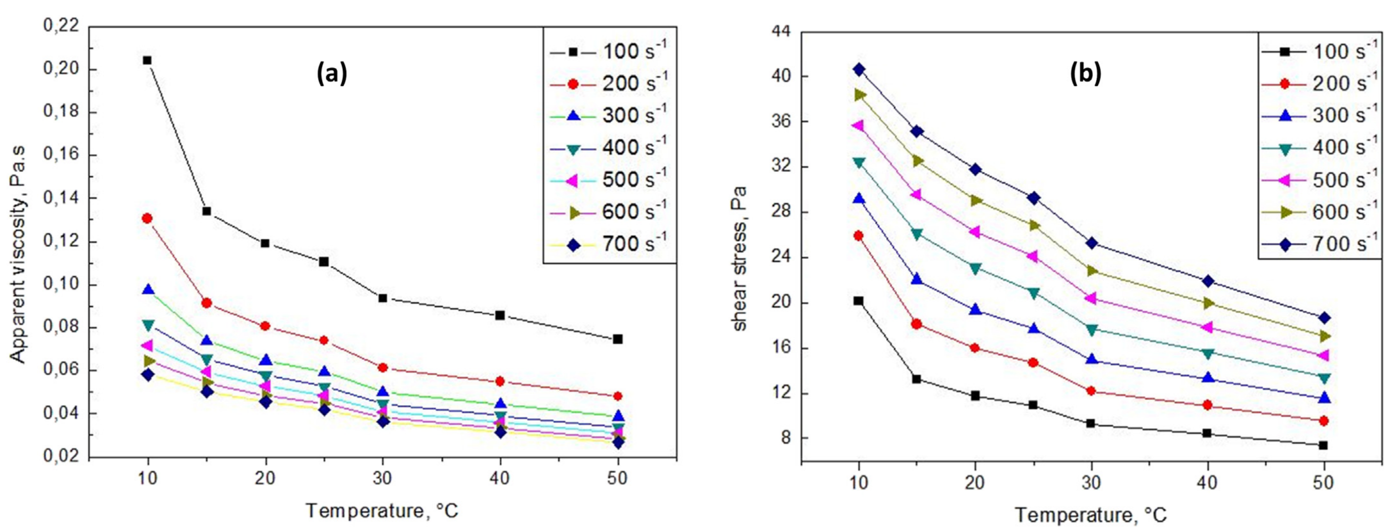

Fig. 2. Temperature effect on the flow properties of sludge at different shear rates: (a) viscosity (b) shear stress.

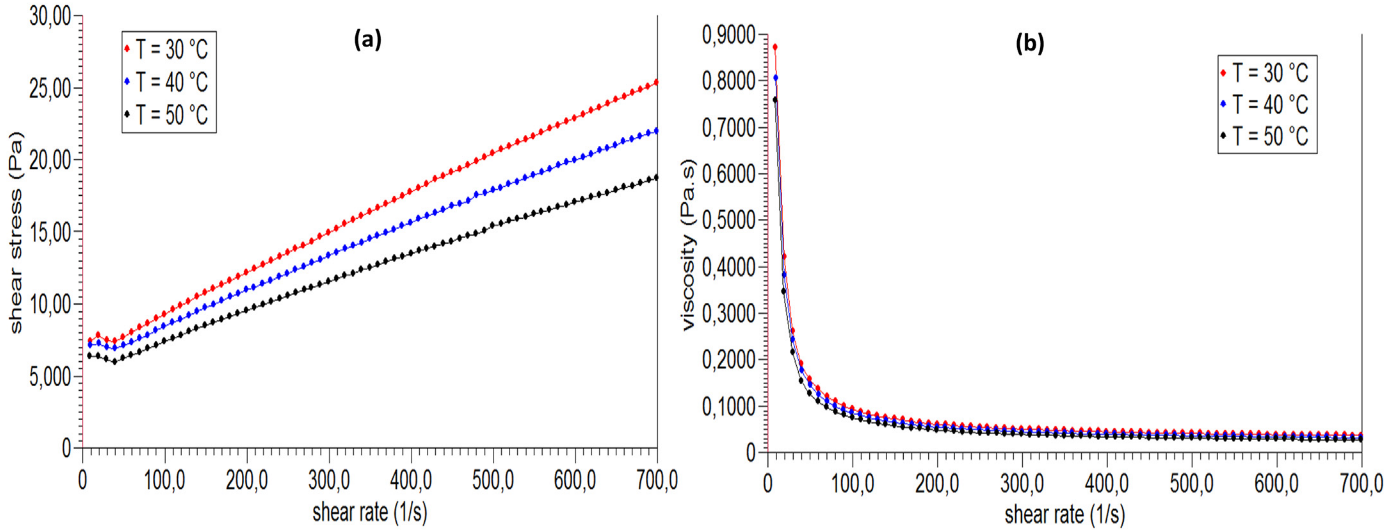

Fig. 3. Flow curves of sludge at different temperatures: (a) shear stress (b) viscosity.

disintegration increases gradually with the shear rate, showing a sharp decrease in viscosity with the increase of the shear rate. When the shear rate increases to a certain extent, the disintegration rate of the structures and the recovery rate of the structures reach a dynamic equilibrium, and the molecules tend to be arranged orderly along the shear direction, and the viscosity gradually becomes constant (Kumar et al., 2014).

In order to model all types of non-Newtonian fluids under different flow conditions, several rheological models have been proposed in the literature. However, some laboratory experiments and field tests of certain fluids require new empirical correlations from curve fitting exercises to describe the non-Newtonian behavior (Sami et al., 2017). Four rheological models, namely the power law model, the Herschel and Bulkley model, the Bingham model and the Casson model, were examined according to Equations (2)–(5) to find the most appropriate relationship to represent the experimental measurements.

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

$$\tau = \tau_0 + K\dot{\gamma}^{n} \quad (3)$$

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

$$\sqrt{\tau} = \sqrt{\tau_0} + \sqrt{\mu\dot{\gamma}} \quad (5)$$

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).

The reliability of each rheological model with the experimental data was evaluated on the basis of the statistical standard error, which is expressed as follows:

$$SE = \left[\frac{\left[\frac{\sum_{i=1}^{n}(x_m - x_c)^2}{n-2}\right]^{\frac{1}{2}}}{X_m^{max} - X_m^{min}}\right] \times 1000 \quad (6)$$

where x_m is the measured value, x_c is the calculated value and n is the number of data points. The experimental data were fitted with the four models as shown in Figs. 4–7 and the results of the modeling analysis are listed in Table 2. From these figures, it was concluded that the Herschel Bulkley model fits the sludge flow behavior very well over the range of shear rate and temperature tested. This was also confirmed by the data in Table 2, where the lowest SE and highest R^2 for the Herschel Bulkley model were reported. The values of the behavior index (n) are less than 1. Therefore, it can be concluded that at different temperatures the fluid exhibits pseudoplastic or shear-thinning behavior.

3.2. Measurement of yield stress

The yield stress is the stress corresponding to the transition from elastic to plastic deformation. When the applied stress is lower, the structural deformation makes the raw sample inherently elastic. However, when the applied stress exceeds the yield point, the deformation causes the sample to flow (Kumar et al., 2017). Experimental application of this definition allows accurate approximations of yield stress to be obtained by (i) graphically or numerically extrapolating shear stress versus shear rate (flow curves) to zero shear rate, and (ii) fitting the experimental data using an appropriate non-linear rheological model for yield stress fluids, such as the Herschel-Bulkley model (Nguyen et al., 2006). The yield stress measurement was performed at different temperatures under controlled shear rate mode. The controlled rate mode is the most common method for measuring yield stress. Fig. 5a–c shows the flow curves obtained for the sludge sample in terms of shear stress and viscosity versus shear rate at different temperatures. The yield stress was measured and compared to that obtained by the most appropriate Herschel Bulkley rheological model in Table 3. In general, the deformation and flow property are governed by the composition of the heavier fractions of the sludge substrate, such as asphaltene, resin. Below the yield point, the fluid cannot move due to the increase in its solid-like (elastic) property, i.e., below the yield point, there is elastic deformation and the applied stress is not sufficient to deform the structural flow and is reversed to its original structure. It was observed in Table 3 that the minimum required stress that allows the sludge to start flowing and deforming its physical structure decreases with increasing temperature, indicating an ease of shear flow of the material by reducing its deformation energy and thus allowing a greater tendency to flow (Sakthivel and Velusamy, 2020). From the comparison of the yield stress results in Table 3, it can be concluded that the yield stress values show good agreement between the experimental values and those predicted by the model.

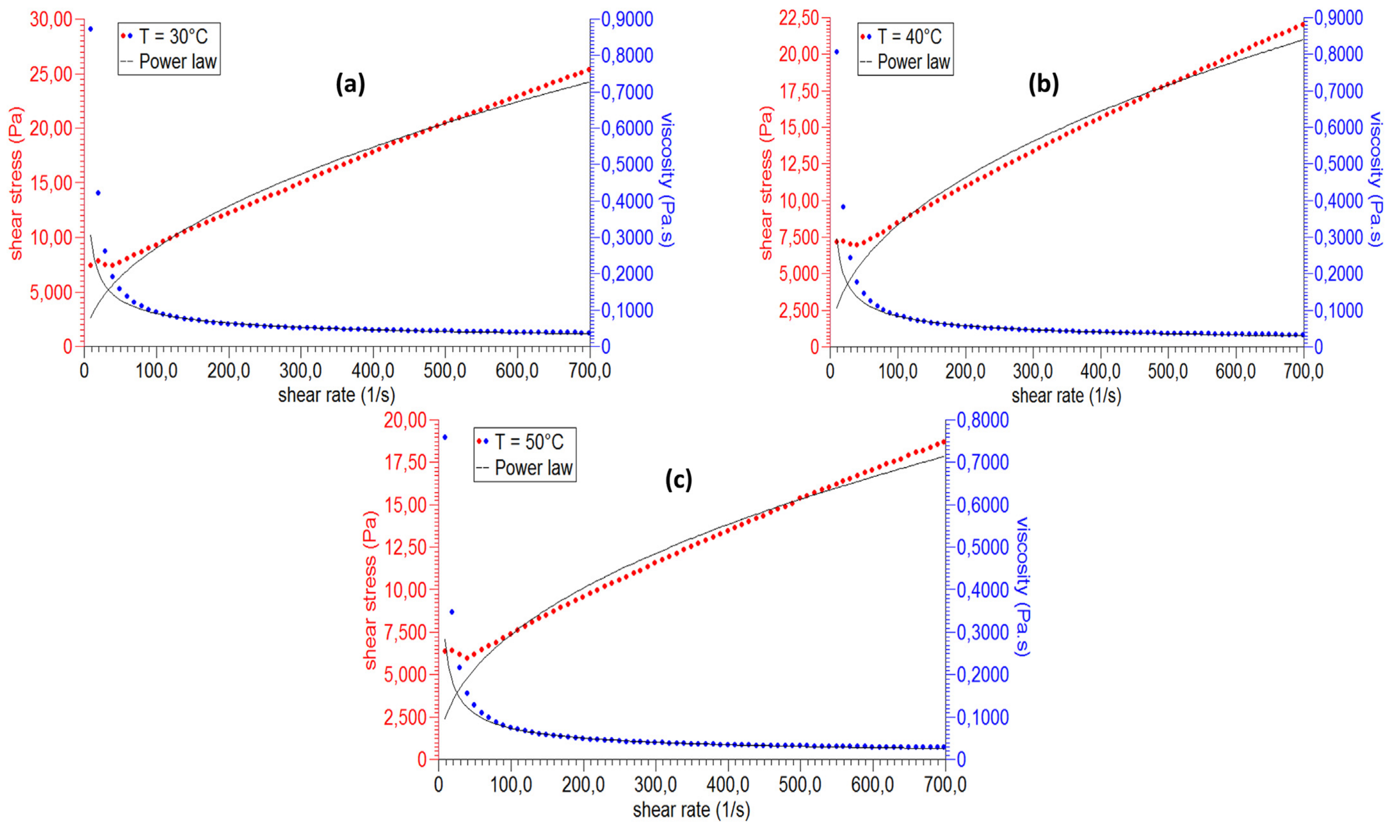

Fig. 4. Modeling the flow behavior of sludge at different temperatures using the Power law model.

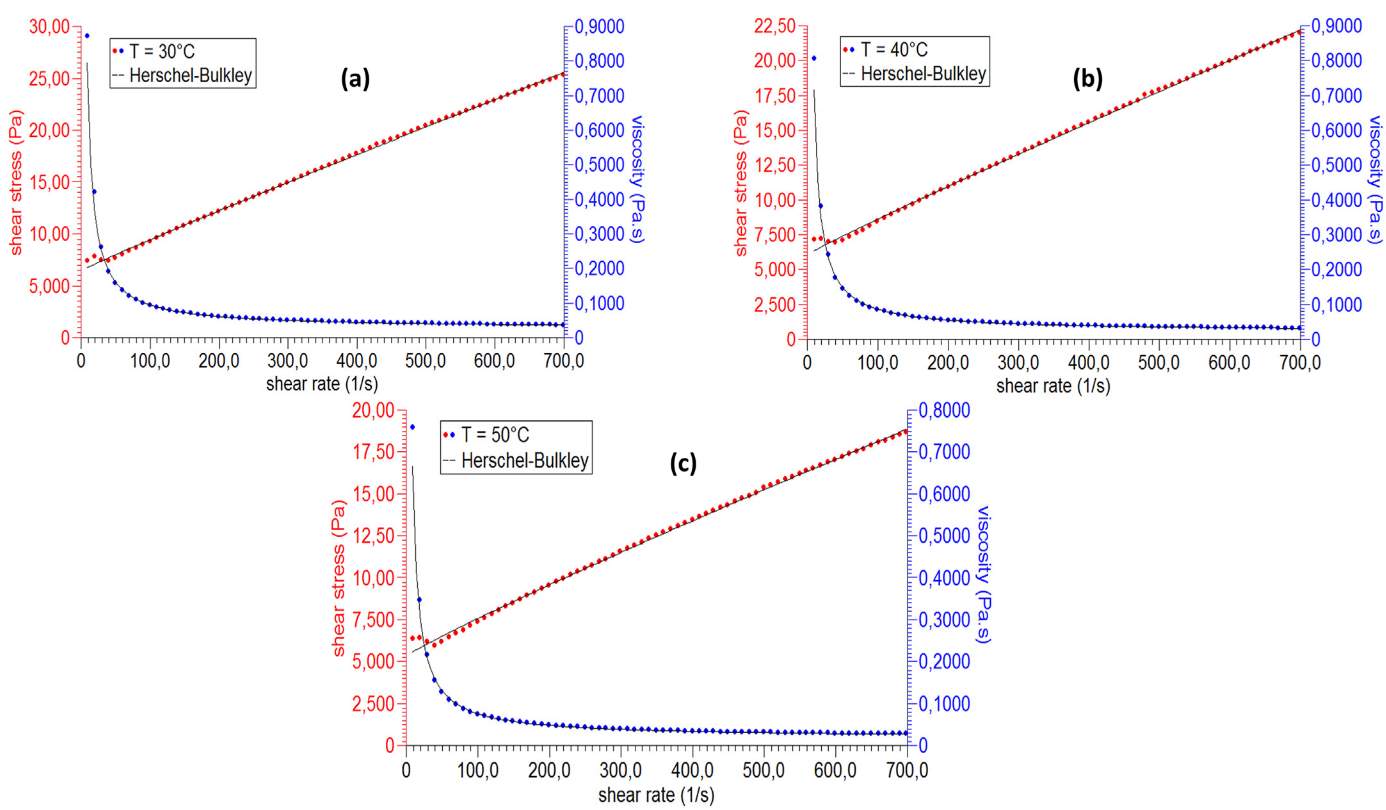

Fig. 5. Modeling the flow behavior of sludge at different temperatures using the Herschel- Bulkley model.

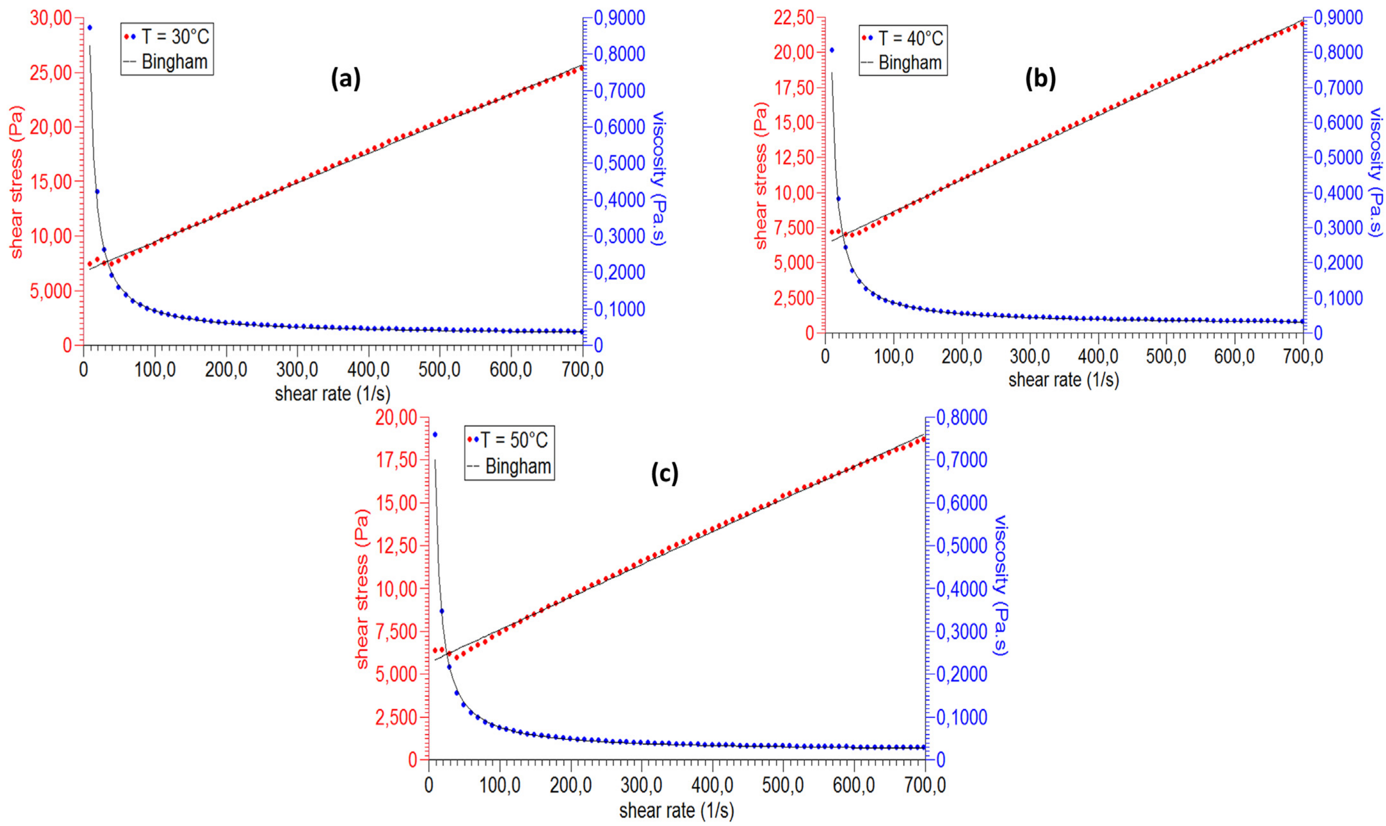

Fig. 6. Modeling the flow behavior of sludge at different temperatures using the Bingham model.

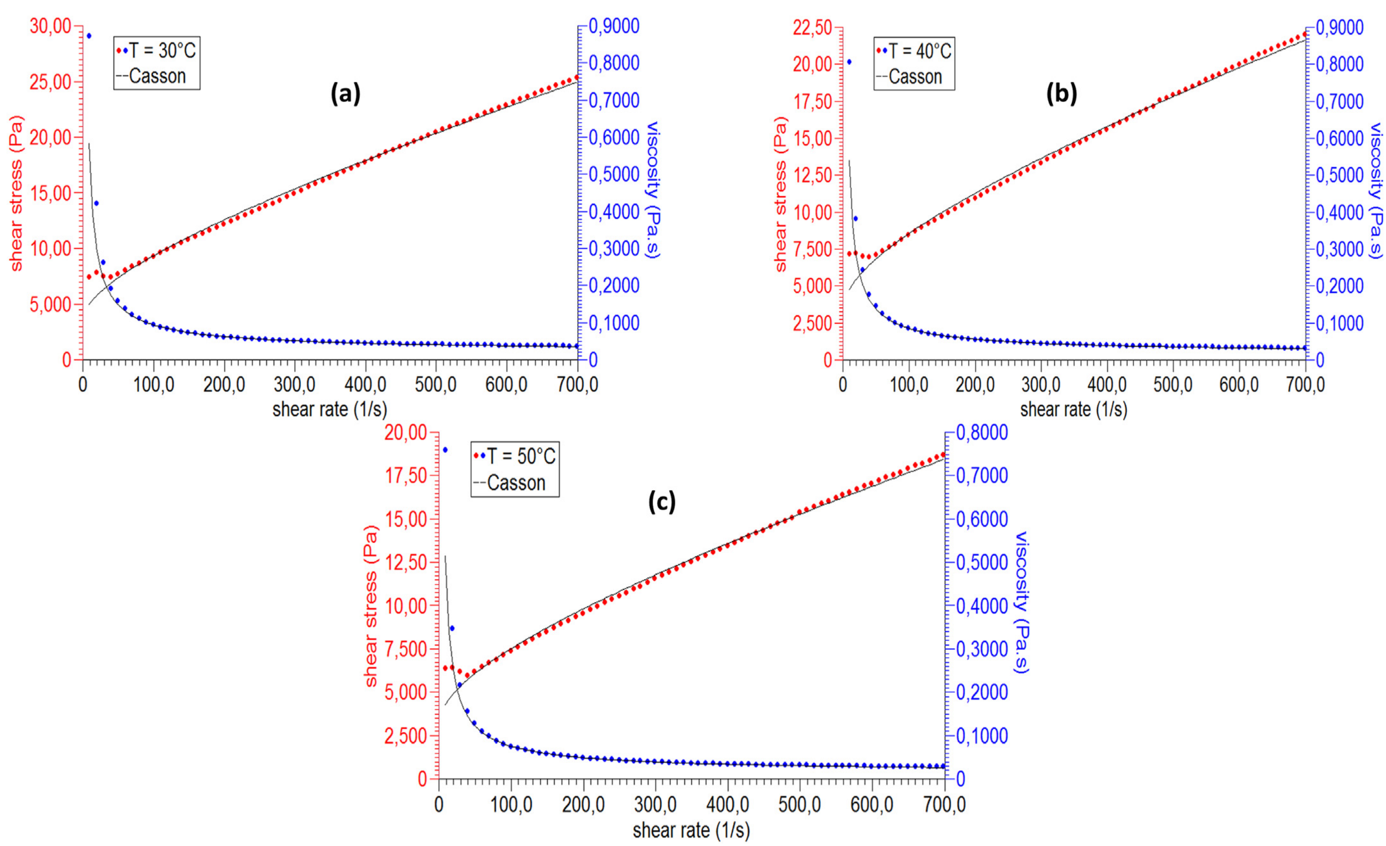

Fig. 7. Modeling the flow behavior of sludge at different temperatures using the Casson model.

Table 2
Rheological modeling analysis of experimental data.

Rheological model	T (°C)	τ_0(Pa)	K (Pa.s^n)	n	μ (Pa.s)	Equations	SE	R^2
Power law	30	–	0.8787	0.5065	–	$\tau = 0.8787.\gamma^{0.5065}$	41.40	0.9837
	40	–	0.9318	0.4756	–	$\tau = 0.9318.\gamma^{0.4756}$	43.09	0.9810
	50	–	0.9007	0.4561	–	$\tau = 0.9007.\gamma^{0.4561}$	43.04	0.9802
Herschel-Bulkley	30	6.459	0.03666	0.9548	–	$\tau = 6.459 + 0.03666.\gamma^{0.9548}$	**6.741**	**0.9995**
	40	6.099	0.03135	0.9528	–	$\tau = 6.099 + 0.03135.\gamma^{0.9528}$	**7.491**	**0.9994**
	50	5.338	0.03111	0.9273	–	$\tau = 5.338 + 0.03111.\gamma^{0.9273}$	**8.634**	**0.9991**
Bingham	30	6.765	–	–	0.0270	$\tau = 6.765 + 0.0270.\gamma$	7.525	0.9994
	40	6.370	–	–	0.0228	$\tau = 6.370 + 0.0228.\gamma$	8.230	0.9993
	50	5.695	–	–	0.01904	$\tau = 5.695 + 0.01904.\gamma$	10.09	0.9988
Casson	30	3.542	–	–	0.0138	$\tau = \left(3.542^{0.5} + (0.0138.\gamma)^{0.5}\right)^2$	19.82	0.9960
	40	3.512	–	–	0.01104	$\tau = \left(3.512^{0.5} + (0.01104.\gamma)^{0.5}\right)^2$	20.55	0.9954
	50	3.241	–	–	8.888 E−3	$\left(3.241^{0.5} + (0.00889.\gamma)^{0.5}\right)^2$	19.96	0.9955

Table 3
Yield stress measurements values and simulation.

Temperature (°C)	Experimental values (Pa)	Yield Stress simulation (Pa)		
		Herschel-Bulkley	Sandard error	R^2
30	7.394	6.459	6.741	0.9995
40	7.132	6.099	7.491	0.9994
50	6.331	5.338	8.634	0.9991

3.3. Dynamic rheology

Dynamic rheology shows the viscoelastic nature of the system under study, as it exhibits comparable viscous and elastic properties. In the linear viscoelastic region (LVR), the measured values of the complex modulus (G*), elastic (storage) modulus (G′), viscous (loss) modulus (G″), and phase angle (δ) truly represent the material properties that are a function of angular frequency (Anto et al., 2020). G* or complex modulus, represents the resistance of the material to the applied strain or the toughness of the material to the applied strain. G′ or elastic modulus, represents the contribution of stress energy that is temporarily stored in the material during the test and can be recovered. G″ or viscous modulus, represents the unrecoverable energy that was required to initiate the flow. δ or phase angle, measures the coupling between the oscillatory input stimulus to the sample response. When δ is equal to 0°, the material behaves as a hookean solid material while when δ is equal to 90°, the material is considered as a Newtonian fluid. When the phase angle δ is equal to 45°, the material's elastic component is equal to its viscous component (G′ = G″) (Taborda et al., 2017).

The following equations were then used to calculate the elastic and viscous moduli from the complex modulus and phase angle:

$$G' = G^* \cos\delta \quad (7)$$

$$G'' = G^* \sin\delta \quad (8)$$

$$G^* = G' + iG'' \quad (9)$$

where

$$\tan\delta = \frac{G''}{G'} \quad (10)$$

The complex modulus versus frequency for the sludge sample at different temperatures (30–50 °C) is shown in Fig. 8. As can be seen, the complex modulus (G*) measured increases progressively with frequency for all different temperatures. The total resistance to applied strain (complex modulus) decreases significantly, which means that the increase in temperature can influence the suspended colloids present in the sludge. The asphaltenes present in the sample are in the form of suspended colloids which are then stabilized by the resins. The formation of clusters between small and large diameter colloidal particles can lead to the formation of aggregates of larger structures. The behavior of sludge as both an elastic and viscous material indicates that the macrostructures created can be affected by heating (Ghannam et al., 2012; Kumar et al., 2017).

In Fig. 9, the frequency dependence profiles of the elastic modulus (G′), viscous modulus (G″) and phase angle δ are presented at the three different temperatures. It was observed that the values of the storage modulus are higher than the corresponding

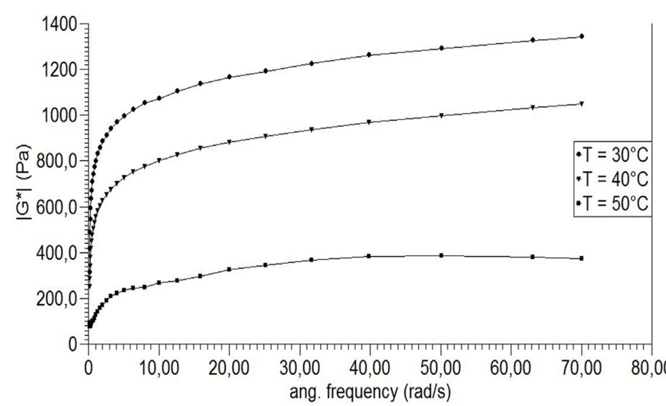

Fig. 8. Complex modulus of sludge at different temperatures.

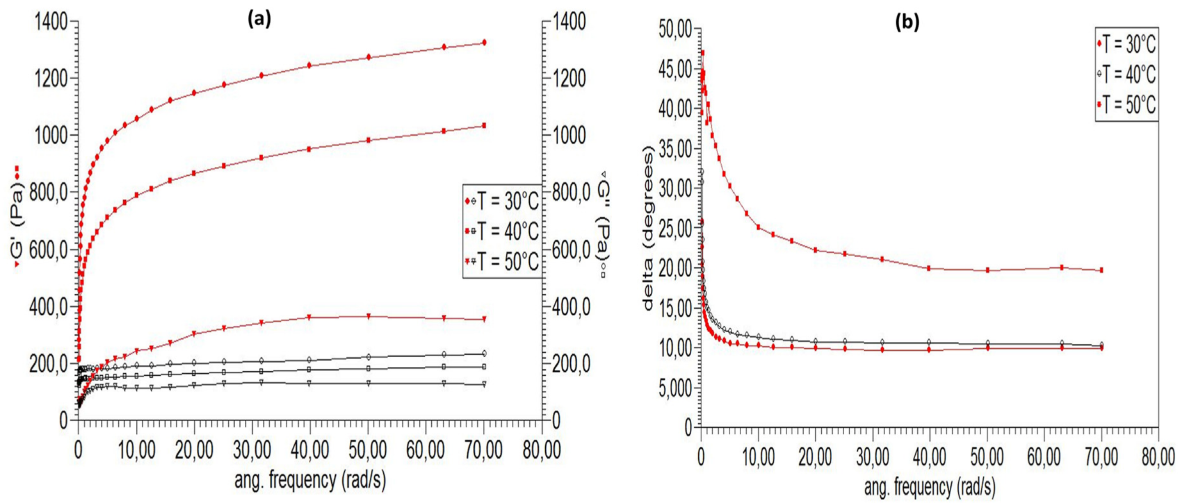

Fig. 9. Storage modulus G′, loss modulus G′ and phase angle δ of sludge as a function of angular frequency at different temperatures.

loss modulus over the range of frequency sweep, indicating that the sludge exhibits highly elastic rather than viscous behavior and that the energy lost per cycle is much lower than the energy stored in the sludge. The phase angle δ is less than 45° (Fig. 9b), confirming that the fluid behavior is dominated by the contribution of the elastic component. Such behavior in the linear viscoelastic region can be strictly related to the aggregation state of asphaltenes and the formation of a three-dimensional network composed of clusters (Taborda et al., 2017). In addition, the higher the solid particle content in the oil sludge, the stronger the colloidal forces and network strength (Liu et al., 2016; Mansur et al., 2015). An increase in temperature leads to a sharp decrease in the values of the viscoelastic moduli (at 50 °C) as expected and an increase in the phase angle δ which is always less than 45° due to structural changes in the viscoelastic network formed, suggesting the same solid-like behavior (Taborda et al., 2017; Sadeghi et al., 2013). Therefore, we can say that throughout the studied experimental condition, the material behaves more like a solid than a liquid.

4. Conclusion

The rheological study of the oil sludge from crude oil storage tank of Algerian refinery led to the following conclusions:

- The rheological behavior of sludge is non-Newtonian shear-thinning or pseudo-plastic at low shear rates where the viscosity decreases with increasing shear rate, and quasi-Newtonian at high shear rates where the viscosity profile becomes independent of shear rate and temperature.
- The increase in temperature had positive effects on the characterization of sludge flow, as it reduced viscosity, shear stress and yield stress.
- Various rheological models are applied to check the best fit to the experimental data. The modeling analysis indicates that the sludge fits the Herschel Bulkley model for all temperatures tested with the lowest standard error and highest correlation coefficient, followed by the Bingham model which also shows a good fit.
- Dynamic rheological measurement indicates that the complex modulus increases with frequency and decreases significantly with temperature, showing structural changes in the viscoelastic network.
- Frequency sweep test revealed that the behavior of the sludge is dominated by the contribution of the elastic component and its energy loss per cycle is much smaller than the stored energy.
- An increase in temperature results in a sharp decrease in the values of viscoelastic moduli, especially elasticity, and a conservation of the same solid-like behavior. Therefore, under all the experimental conditions studied, the material behaves more like a solid than a liquid.

Declaration of competing interest

The author declares that there is no conflict of interest regarding the publication of this article.

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