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Improvement of Flow Properties of Highly Waxy Crude Oil with the application of SiO_2 and Graphene Oxide nanoparticles aided by Light Crude Oil

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Abstract: The rheological behaviour of crude oil mixtures comprising both heavy and light components along with chemical additives is of significant interest due to its relevance in the petroleum industry. Understanding the flow properties of these mixtures is crucial for efficient transportation, processing, and refining operations. This study investigates the effectiveness of incorporating light crude oil and nanoparticles as additives to mitigate wax-related flow issues. The test involves six different waxy/heavy and light crude oil mixture concentrations along with Graphene Oxide and Silicon Dioxide nanoparticles. Rheological tests were conducted to evaluate the impact of these additives on the flow behaviour of waxy crude oil. The rheological characterization involved measuring the viscosity, yield stress, and shear rate dependencies of waxy crude oil samples with varying concentrations of light crude oil and nanoparticles. Nano-materials have emerged as promising candidates for mitigating wax deposition issues and enhancing the fluidity of crude oil. Nevertheless, the intricate nature of crude oil has made it challenging to elucidate the underlying mechanisms governing wax resolution, crystal modification, and flow improvement. The results revealed notable improvements in the rheological properties of the treated crude oil, including reduced viscosity and enhanced flowability, even at low temperatures. The incorporation of light crude oil and nanoparticles proved to be a promising strategy for modifying the rheological behaviour of waxy crude oil, ensuring improved flow assurance. This study contributes to understanding innovative approaches to address flow challenges in waxy crude oil systems, thereby offering potential solutions for the oil and gas industry.

Introduction

Petroleum is a naturally occurring fossil fuel formed from the decay of organic matter over millions of years. It is a complex mixture of hydrocarbons, primarily consisting of carbon and hydrogen atoms, along with smaller amounts of sulfur, nitrogen, and other elements (Li et al., 2023). Petroleum is found in underground reservoirs and is extracted through drilling operations (Khalilidermani, 2023). It serves as a vital energy resource, powering transportation, industry and heating systems worldwide (Ahmad and Zhang, 2020). It is also refined into various products such as gasoline, diesel, jet fuel, and petrochemicals, which are essential for modern life (Chakrovorty et al., 2021). The motivation behind studying and improving flow properties in waxy crude oil lies in the practical challenges associated with its transportation and processing. Crude oil extracted from reservoirs often contains waxes, which are solid hydrocarbons. These waxes can solidify and precipitate out of the oil under certain conditions such as low temperatures or changes in pressure, forming a viscous and sticky mixture that hampers the flow of oil through pipelines and equipment (Qiu et al., 2023). It directly addresses transportation, processing and overall operational efficiency challenges. By enhancing flow properties, the industry can ensure smoother and more reliable transportation of crude oil through pipelines and equipment, reducing the risk of blockages and associated

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downtime. This optimization also leads to decreased energy consumption, as less energy is required to overcome flow restrictions caused by waxes. A wide range of qualities, from light, low-viscosity oils to heavy, very viscous ones, are produced by various crude oil sources (Hasan et al., 2010; Souas et al., 2020). These composition and rheological behavior variations for the petroleum business bring special challenges and opportunities. The rheological properties of crude oil mix constituting both heavy and light components have drawn growing interest due to their significance in many phases of oil production, transportation and refining operations (Zhang et al., 2017). Crude oils' rheological characteristics are quite helpful for any procedures involving the transportation of fluids from one place to another. High-viscosity oils may have difficulty flowing through the porous rock formation of the reservoir (Wang and Sheng, 2017), leading to reduced production rates and increased pressure drop. In such cases, measures like thermal recovery methods or adding viscosity-reducing agents (e.g., solvents or surfactants) might be employed to improve production efficiency. One approach to reducing the viscosity of heavy crude oil is to form oil-in-water emulsions (Alba and Jorge, 2015; Bu-Ali et al., 2021). In this process, water is mixed with crude oil to create tiny water droplets dispersed throughout the oil phase. The emulsification reduces the overall viscosity of the mixture, making it easier to transport through pipelines (Dong et al., 2022). Emulsification can be achieved using mechanical means or by adding surfactants or emulsifying agents to stabilize the emulsion (Oil-in-water Emulsions, 2023). Heating heavy crude oil is another technique employed to reduce viscosity for pipeline transportation. Increasing the crude oil's temperature makes its molecular interactions weaken, leading to a decrease in viscosity (Zhu et al., 2021). Heat can be applied at different stages of the transportation process, such as preheating at the source, in-transit heating, or heating at the destination (Mun et al., 2022). Proper temperature control is crucial to avoid issues like thermal degradation or excessive energy consumption (Li et al., 2020). It is essential to mention that each of these techniques has its advantages and limitations, and their effectiveness may vary depending on the specific characteristics of the crude oil and the transportation conditions (Flowlines, 2021). Also, choosing a particular method may depend on economic considerations and environmental factors to ensure sustainable and efficient transportation of crude oil. Researchers continue exploring new technologies and improvements in existing methods to address the challenges of transporting heavy crude oil through pipelines (Oil Emulsions, 2019). Blending heavy crude oils with lighter oils, hydrocarbon gases, or alcohols is another effective approach to reduce the viscosity of heavy oils (Ramírez-de-Santiago et al., 2023). This technique is commonly used in the petroleum industry to improve the flow properties of heavy crude oils during transportation, refining, and processing. The process involves mixing heavy crude oil with lighter hydrocarbons or alcohols, which results in a reduction of the overall viscosity of the blend. This allows for easier handling and transportation through pipelines, as well as better processing in refining units. Blending offers several advantages as a viscosity reduction technique. It is a flexible and controllable process that allows for customization of the blend's properties to meet specific transportation and refining requirements. Additionally, blending is often more cost-effective compared to other viscosity reduction methods, such as heating or the use of chemical additives. However, it is essential to carefully consider the compatibility of the blend components and their potential impact on the final product's quality. This study aims to provide an overview of heavy crude oil's rheological characteristics and nanoparticles' emerging role in modifying its rheological behavior. It will discuss the synergistic effects that nanoparticles can have when combined with waxy and heavy crude oil mixtures, potentially revolutionizing the energy industry's approach to handling and optimizing the utilization of these valuable resources.

Materials and Method

For the current investigation, the two crude oil samples (One waxy/heavy oil and one light crude oil) were provided by Oil India Limited (OIL) in India’s Northeastern oil fields. Silicon Dioxide was purchased from a chemical shop, and graphene oxide was synthesized using the improved Hammer method. Experimental and analytical approaches are used to determine the characteristics of crude oil in accordance with national and international standards. ASTM Standard D86-08a and ASTM Standard D97-08, were employed to measure density and determine the pour point of crude oil. The API gravity was determined using the API formula i.e., API gravity = (141.5/S.G.) – 131.5 (here, S.G.= Specific Gravity). The paraffin wax content of crude oil was calculated in accordance with ASTM Standard UOP46-85 and the water content was determined using ASTM D 96-58 T (Singh, 2021). The SARA distribution of crude oil (i.e., Saturates, Aromatics, Resins, and Asphaltenes) was tested with

ASTM D2007-93 procedure. The rheological studies were done with the help of Anton Paar Rheometer MCR-72 model.

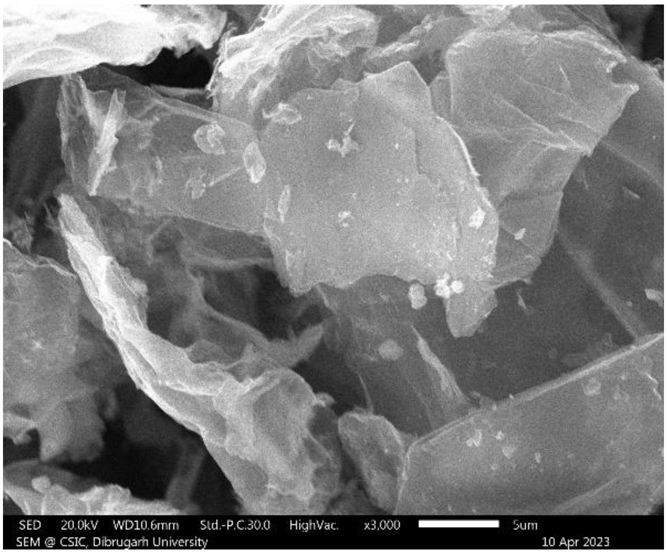

Figure 1. SEM images of GO nanoparticles.

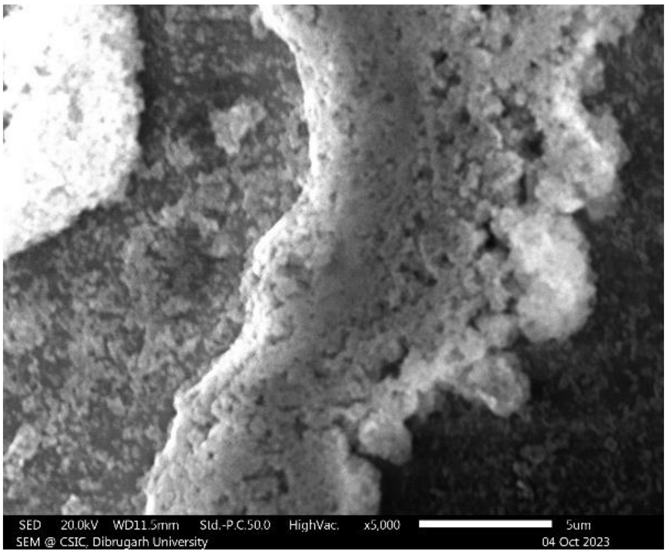

Figure 2. SEM images of SiO_2 nanoparticles.

Figure 1 displays a scanning electron microscopy (SEM) image of graphene oxide (GO) sheets. These sheets are depicted as thin, randomly clustered structures characterized by clear edges, wrinkled surfaces, and folds. On average, the dimensions of these sheets measured approximately 10 x 20 micrometers.

Figure 2 displays the SEM image of SiO_2 (silicon dioxide) nanoparticles reveals a fascinating microscale landscape. The nanoparticles appear as small, spherical entities uniformly dispersed across the image. Each nanoparticle exhibits a smooth and spherical surface, indicative of its well-defined and compact structure. Their sizes typically range in the nanometer scale, making them suitable for various applications. The SEM image provides a detailed view of the individual nanoparticles, showcasing their uniformity and size distribution.

Results and Discussion

The Standard ASTM methods were used to determine the properties and composition of the crude oil samples (Jha et al., 2014).

Waxy crude oil was blended with the light crude oil and nanoparticles in 5 different concentrations of the Waxy Crude (WC), light Crude (LC) and nanoparticles. The nanoparticles used were Graphene Oxide (GO) and Silicon Dioxide (SiO_2) both with different concentrations of 0.5 % and 1% separately.

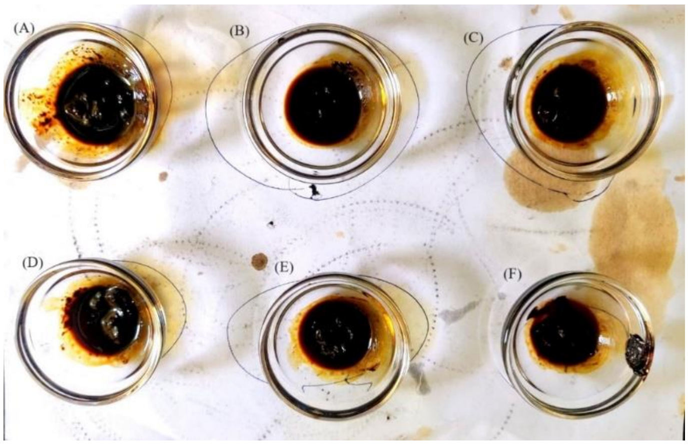

Figure 3. (A) WC ;(B) WC+LC20% ;(C) WC+LC20%+GO(0.5%) ;(D) WC+LC20%+GO(1%) ;(E) WC+LC20%+SiO_2(0.5%) ;(F) WC+LC20%+SiO_2(1%).

The rheological measurements were carried out using the Anton Paar mcr-72 rheometer. All measurements except the viscosity vs temperature graphs were carried out at a room temperature of 27°C. This rheometer has several operation test modes for performing various tests, such as viscosity, yield stress, amplitude sweep, frequency sweep etc.

Both SiO_2 and GO nanoparticles possess high surface area and can adsorb onto the surface of wax molecules due to their interactions, such as van der Waals forces and hydrogen bonding (Biswas et al., 2022; Yang et al., 2023; Taheriazam et al., 2023; Mandal et al., 2024). This adsorption alters the surface properties of the wax, leading to changes in its rheological behavior, crystallization kinetics, and thermal properties. For instance, nanoparticles can inhibit the growth of wax crystals or modify their morphology, resulting in finer and more dispersed crystal structures. Also they act as heterogeneous nucleation sites, promoting the nucleation of wax crystals and reducing the supercooling required for crystallization. By providing nucleation sites, nanoparticles facilitate the formation of smaller and more uniformly distributed wax crystals, which can impede the coalescence and growth of crystals. This mechanism ultimately leads to the inhibition of macroscopic wax deposition and the mitigation of flow assurance issues.

Table 1. The Properties and composition of the heavy and light crude oils.

Parameters	Waxy/Heavy Crude Oil	Light Crude Oil
Specific Gravity	0.90411 gm/ml^3	0.88785 gm/ml^3
API Gravity	25.21	41
Pour Point	30^{0}C	12^{0}C
Water Content	0.7	Nil
WAX (wt%)	7.8	7.3
Saturates(wt%)	65.66	66.2
Aromatics(wt%)	20.4	22.2
Resins(wt%)	11.12	10.5
Asphaltenes(wt%)	0.35	0.29

Table 2. Samples and their compositions.

Sl no.	Sample	Concentration
1	A	Waxy/Heavy Crude Oil (WC)(100%)
2	B	WC+ Light Crude(LC)(20%)
3	C	WC+LC20%+Graphene Oxide (GO)(0.5%)
4	D	WC+LC20%+Silicon Dioxide (SiO_2)(0.5%)
5	E	WC+LC20%+GO(1%)
6	F	WC+LC20%+SiO_2(1%)

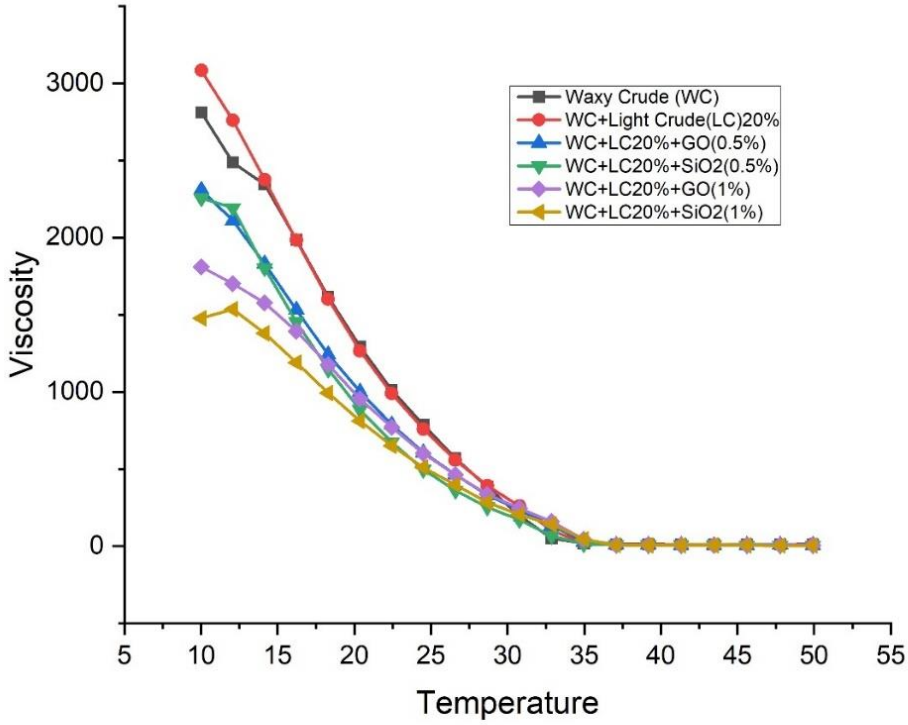

Figure 4. Viscosity vs temperature profile for all mixtures.

Viscosity vs Temperature Profile

The viscosity of crude oil is an essential physical property that affects its flow behavior during transportation, refining, and processing (Luo et al., 2022). Generally, crude oils can be categorized into heavy and light crude oils based on their API gravity (Mohammadi et al., 2020). Heavy crude oils have a lower API gravity (below 20° API) and are more viscous, while light crude oils have a higher API gravity (above 20° API) and are less viscous (Niyonsaba et al., 2019). The viscosity of crude oil varies significantly with temperature (Al-jabri et al., 2021). As temperature increases, the viscosity of both heavy and light crude oils decreases, making them easier to flow. However, the rate of viscosity reduction is different for heavy and light crude oil mixtures (Vanini et al., 2020).

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Heavy crude oils have higher molecular weight and contain more complex hydrocarbons, contributing to their higher viscosity than light crude oils (Tirado et al., 2023). The viscosity vs. temperature profile for a heavy crude oil mixture typically shows a non-linear relationship,

characterized by a steeper decrease in viscosity at higher temperatures. The viscosity remains relatively high at lower temperatures, making the oil more challenging to pump and transport.

Light crude oils have lower molecular weight and contain simpler hydrocarbons, resulting in lower viscosity compared to heavy crude oils. The viscosity vs. temperature profile for light crude oil shows a more gradual decrease in viscosity with increasing temperature. Light crude oils are generally easier to pump and flow at lower temperatures but can become less viscous at higher temperatures (Yao et al., 2023).

It's important to note that the viscosity-temperature relationship is not uniform for all crude oil samples as it depends on the specific composition of the mixture (Gao et al., 2023). Also, various heavy and light crude oil blends exist, and the viscosity behavior can differ between different oilfields and regions.

To get a precise viscosity vs. temperature profile for a specific heavy and light crude oil mixture, laboratory tests were carried out (fig. 4). These tests show a gradual improvement in the viscosity of crude oil as the percentage of light crude oil was increased.

Viscosity vs Shear Rate profile

The viscosity vs. shear rate profile for heavy and light crude oil mixtures will typically exhibit different behaviors due to their varying molecular compositions (fig.5) (M'barki et al., 2023). Heavy crude oil usually has a higher viscosity at low shear rates and exhibits a more significant increase in viscosity with increasing shear rates (Wang et al., 2022). On the other hand, light crude oil tends to have lower viscosity at low shear rates and may show a more gradual increase in viscosity as the shear rate increases. The exact profiles will depend on the mixture's specific blend ratio and temperature conditions. For the current experiments, the temperature used was room temperature of 27°C.

Shear Stress vs Shear Rate Profile

The relationship between shear stress and shear rate for heavy crude oil can often show a non-linear and more pronounced increase in shear stress with increasing shear rate (fig.6). This is known as "shear-thinning" behavior, where the viscosity decreases as the shear rate increases (Zhao et al., 2022). The curve might be steep at low shear rates and gradually flatten as shear rates increase further. Few of the shear rate and shear stress values are shown in the table below (table 3).

Table 3. Shear rate and shear stress values.

Sample Composition \ Shear Rate	0.141 [1/s] (Shear Stress)	8.91 [1/s] (Shear Stress)	100 [1/s] (Shear Stress)
Waxy Crude (WC)	12.627 Pa	39.544 Pa	57.422 Pa
WC+ Light Crude (LC) 20%	22.393 Pa	27.25 Pa	43.518 Pa
WC+LC20%+GO (0.5%)	15.822 Pa	17.203 Pa	31.474 Pa
WC+LC20%+SiO_2(0.5%)	7.2172 Pa	9.7706 Pa	22.152 Pa
WC+LC20%+GO (1%)	4.6154 Pa	6.862 Pa	15.695 Pa
WC+LC20%+SiO_2(1%)	1.8612 Pa	3.986 Pa	12.155 Pa

Heavy crude oil is mainly composed of larger, more complex hydrocarbon molecules, which tend to interact more strongly and resist flow, giving us higher viscosity and density than light crude oils. The relationship between shear stress and shear rate for light crude oil might exhibit a more linear and proportional increase in shear stress with shear rate. This is closer to "Newtonian" behavior, where the viscosity remains relatively constant regardless of shear rate (Fakroun and Benkreira, 2019).

Storage modulus and Loss modulus

The storage modulus and loss modulus are key rheology parameters that describe materials' viscoelastic behavior, including crude oil mixtures. The storage modulus (G') represents the energy stored in a material during deformation, while the loss modulus (G") indicates the energy dissipated as heat during deformation (Bavoh et al., 2020).

The storage modulus and loss modulus profiles will differ for all the crude oil mixtures due to their distinct molecular compositions and viscoelastic properties (Gorbacheva and Ilyin, 2021). Heavy crude oil mixtures generally tend to have higher storage and loss moduli than light crude oil with nanoparticle mixtures. This indicates that heavy crude oil mixtures store and dissipate more energy during deformation, making them more viscoelastic than their lighter counterparts (fig.7).

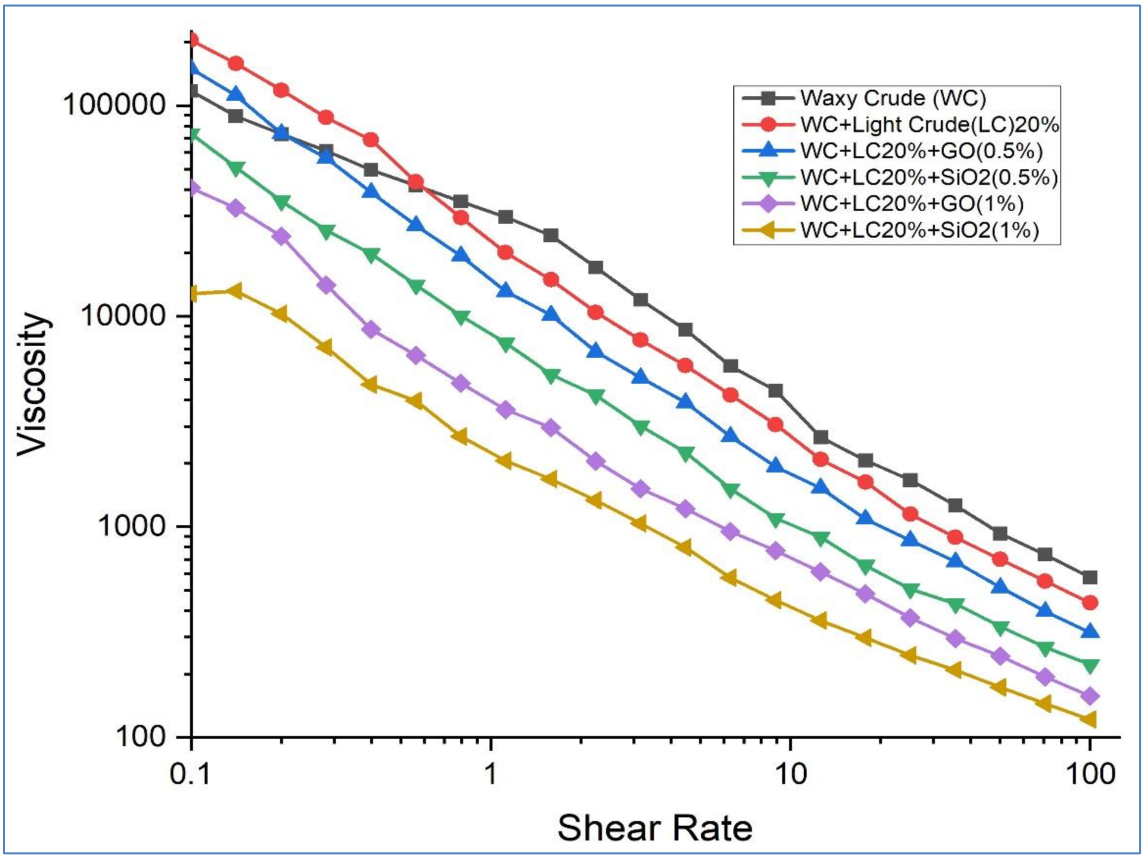

Figure 5. Viscosity Vs Shear rate profile for all mixtures.

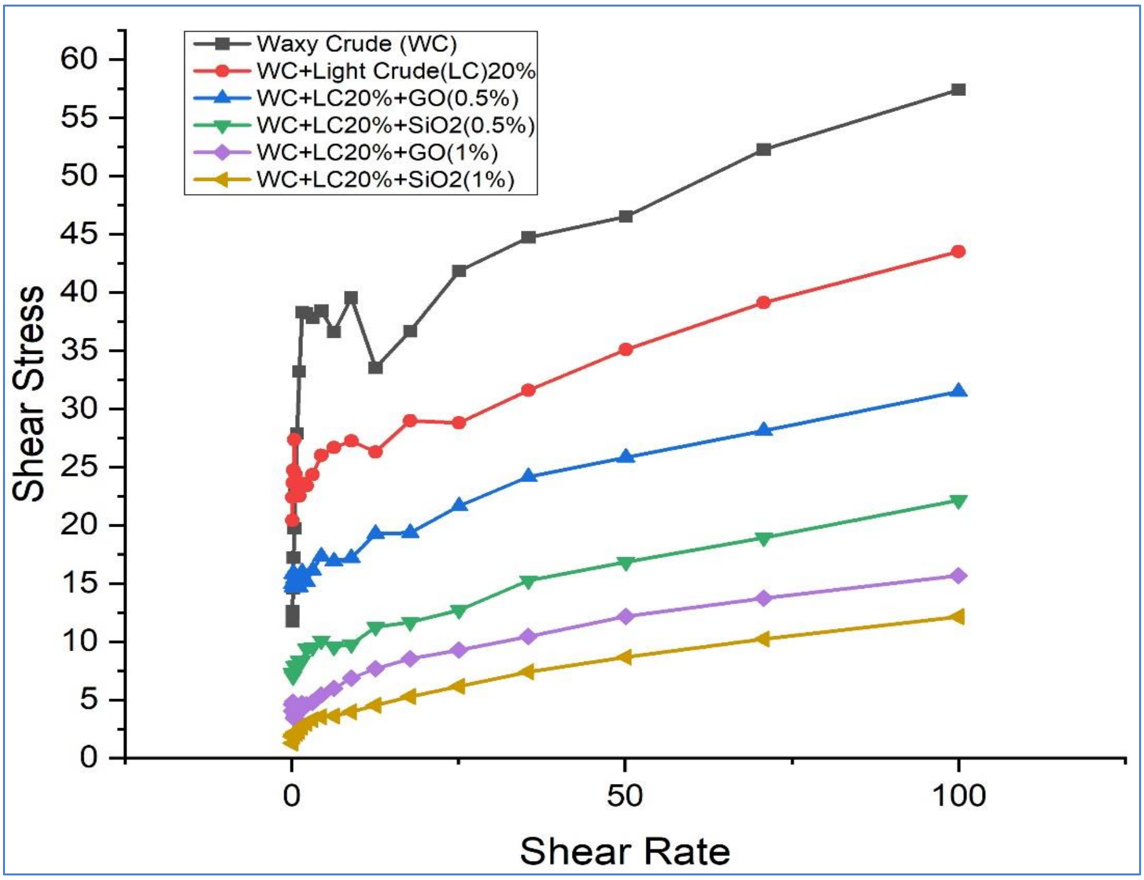

Figure 6. Shear Stress vs Shear Rate Profile for all mixtures.

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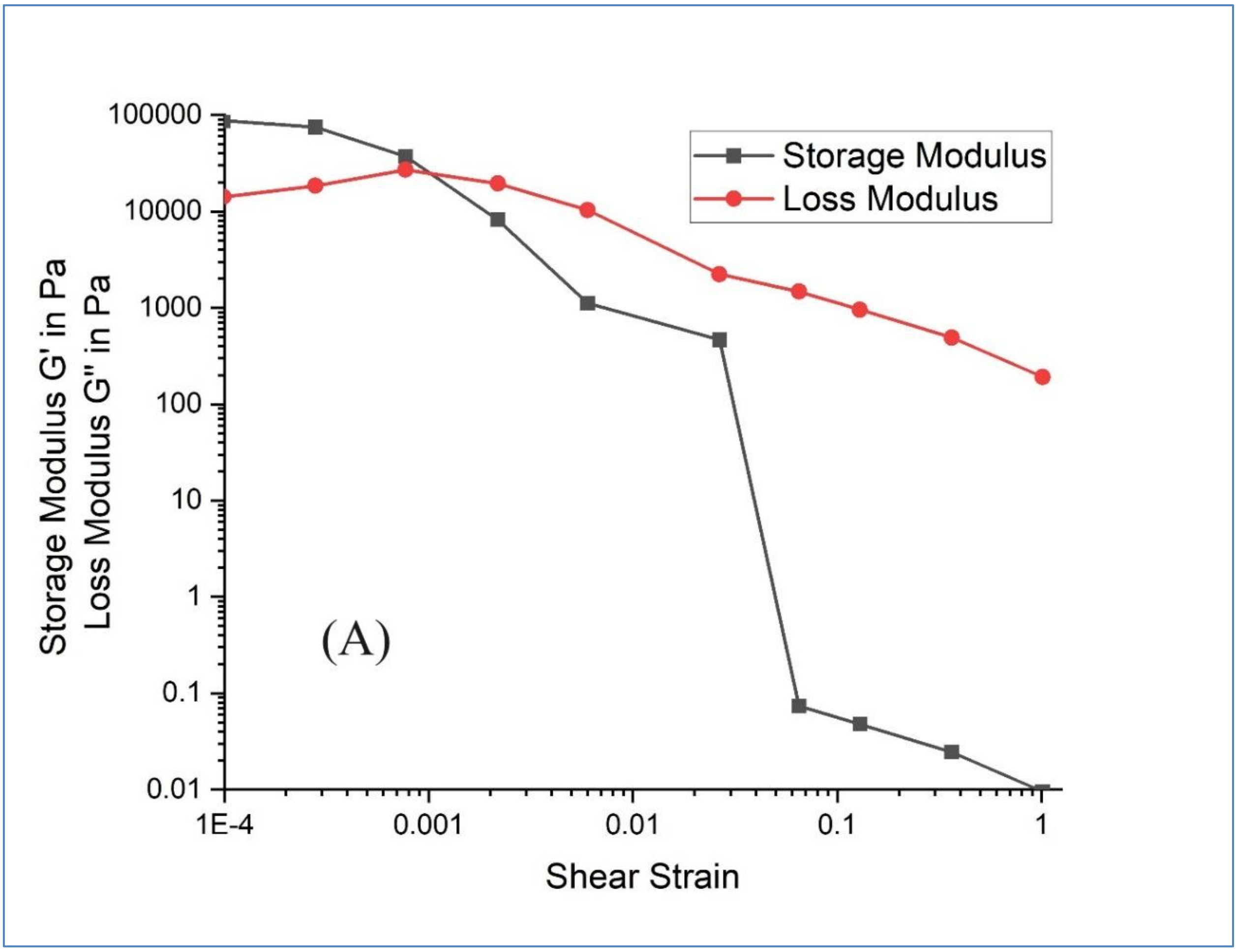
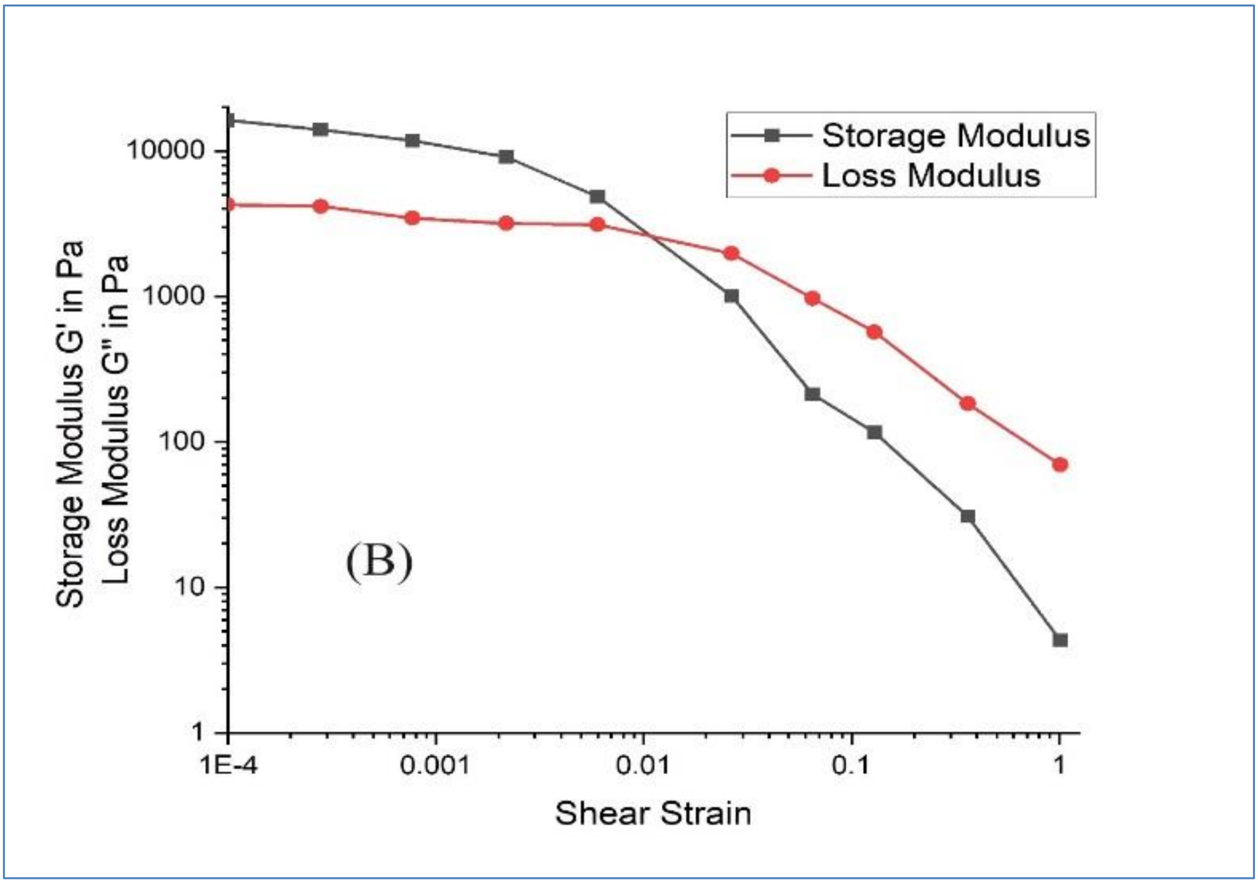

Storage Modulus
Loss Modulus
Storage Modulus G' in Pa
Loss Modulus G'' in Pa
100000
10000
1000
100
10
1
0.1
0.01
(A)
1E-4
0.001
0.01
0.1
1
Shear Strain


Storage Modulus
Loss Modulus
Storage Modulus G' in Pa
Loss Modulus G'' in Pa
10000
1000
100
10
1
(B)
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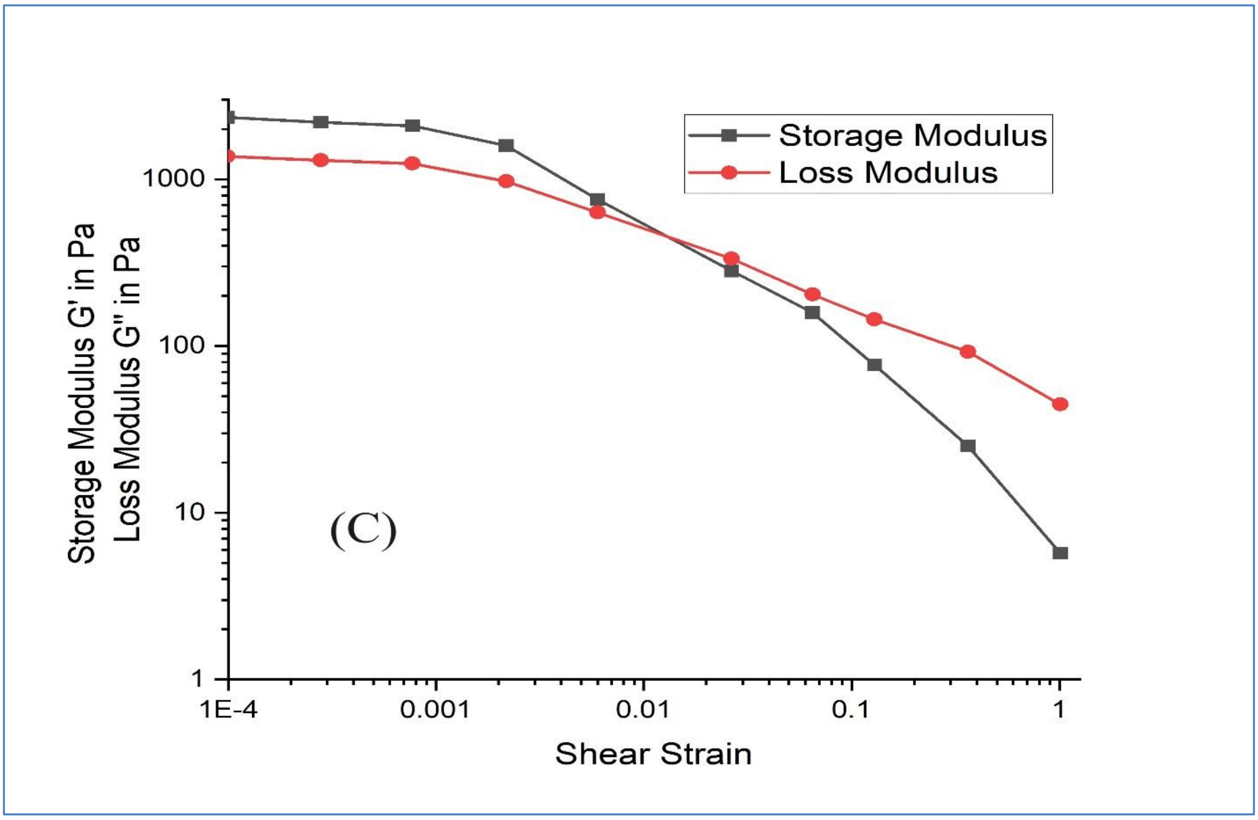
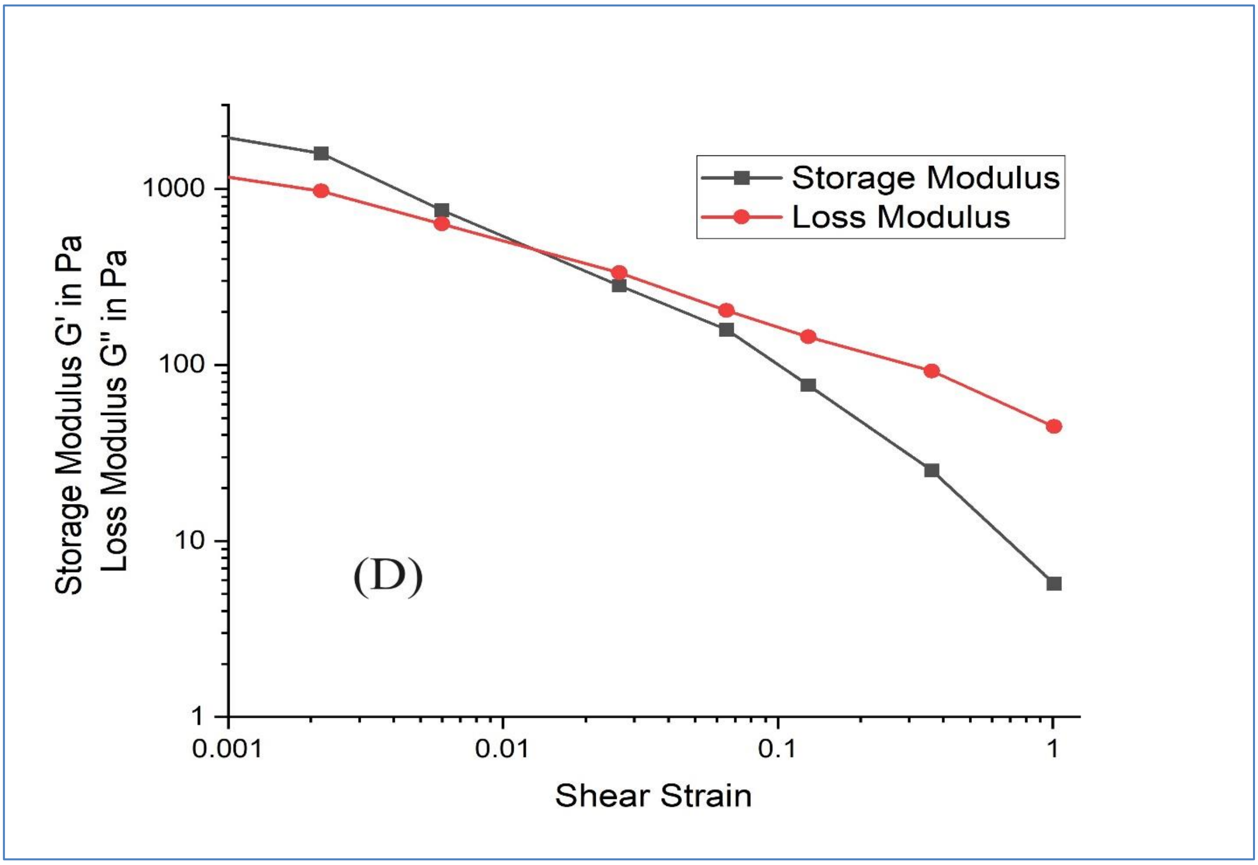
0.1
1
Shear Strain





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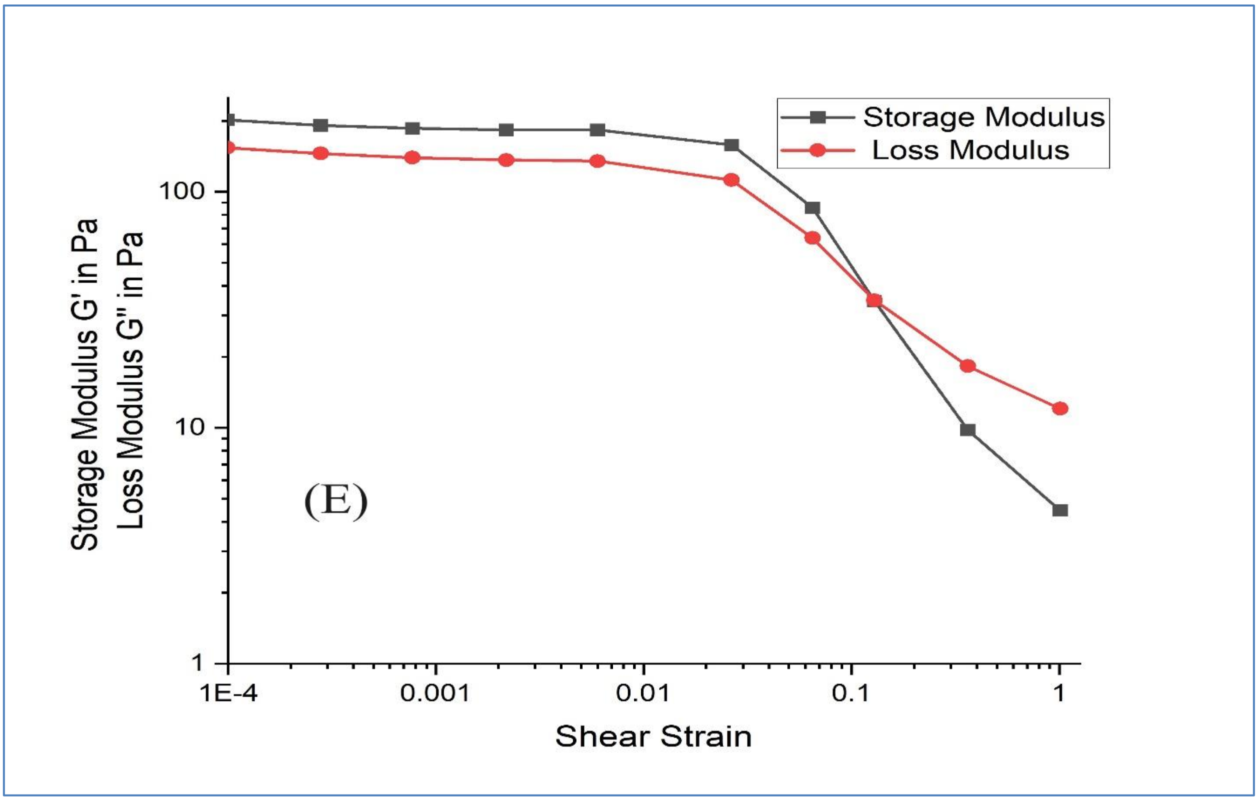

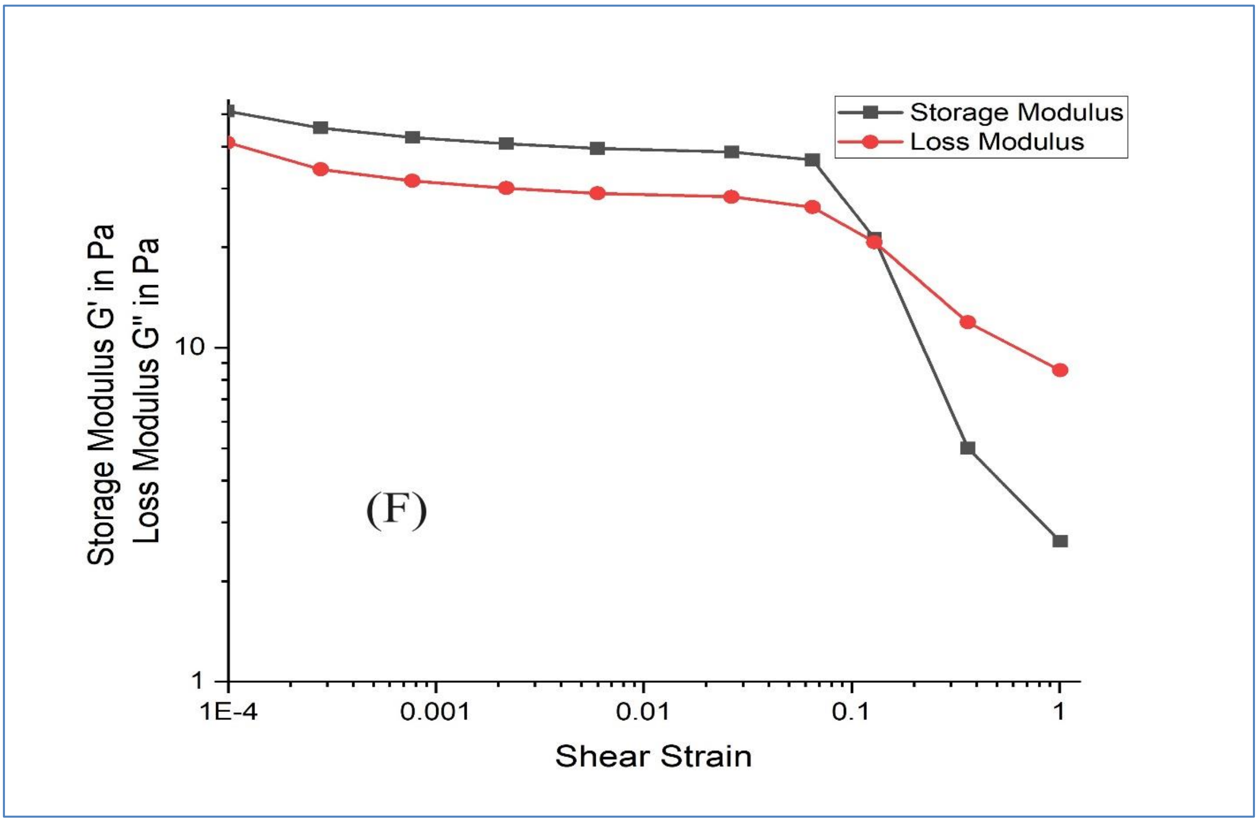

Figure 7. Storage and Loss modulus vs Shear Strain for (A) WC ;(B) WC+LC20% ;(C) WC+LC20%+GO (0.5%);(D) WC+LC20%+SiO_2(0.5%);(E) WC+LC20%+GO (1%) ;(F) WC+LC20%+SiO_2(1%).

Conclusion

In conclusion, the rheological tests on waxy/heavy and light crude oil mixtures with nanoparticles have provided valuable insights into their viscoelastic behavior. The viscosity vs. shear rate profiles revealed distinct characteristics for all types of crude oil mixtures. Waxy/Heavy crude oil mixtures displayed higher viscosities at low shear rates, with a more pronounced increase as the shear rate increased i.e., 573.88 mPa-s at 100 s^{-1}. Conversely, light crude oil and nanoparticle mixtures exhibited lower viscosities at low shear rates, with a more gradual increase in viscosity with higher shear rates i.e., 59.382 mPa-s for WC+LC20%+SiO_2(1%) and 121.37 mPa-s for WC+LC20%+GO (1%) both at 100 s^{-1}. Moreover, the shear stress vs. shear rate profiles showed that waxy/heavy crude oil mixtures had higher shear stresses at a given shear rate i.e., 57.422 Pa at 100 s^{-1} compared to light crude oil mixtures with nanoparticles i.e., 12.155 Pa for WC+LC20%+SiO_2(1%) and 15.695 Pa for WC+LC20%+GO (1%) both at 100 s^{-1}, indicating a more viscoelastic nature. The storage modulus and loss modulus data further supported these observations. Waxy/Heavy crude oil mixtures exhibited higher storage and loss moduli, highlighting their greater ability to store and dissipate energy during deformation. The findings emphasize the significant influence of crude oil composition and concentration on its rheological properties. Understanding these differences is vital for optimizing transportation and processing strategies in the oil and gas industry. While the study on enhancing flow properties of highly waxy crude oil using SiO_2 and Graphene Oxide nanoparticles aided by light crude oil presents promising findings but has limitations. These include the narrow scope of the study, which may not encompass all influencing factors in real-world scenarios, and the lack of real-world applicability due to laboratory conditions. The conditions in a controlled laboratory environment can not fully replicate the complexities of actual oil production and transportation processes, where factors such as pipeline length, varying temperatures, and the presence of impurities can influence flow behavior differently. Despite its insights, further research is needed to address these limitations and provide a more comprehensive understanding of effective flow enhancement strategies for waxy crude oils. Future research could involve conducting field studies or implementing advanced simulation techniques better to simulate the complexities of oil production and transportation processes. This could include factors such as pipeline length, flow dynamics, and environmental conditions. Overall, the results obtained from these rheological tests provide valuable data for engineers and researchers in the oil and gas sector to make informed decisions regarding the handling, transportation, and processing of waxy/heavy, light crude oil and nanoparticle mixtures.

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Conflict of Interest

The authors declare no conflict of interest.

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