

# Influence of Varying Oil–Water Contents on the Formation of Crude Oil Emulsion and Its Demulsification by a Lab-Grown Nonionic Demulsifier

Ahmad A. Adewunmi, Syed Muhammad Shakil Hussain,\* Shirish Patil, and Muhammad Shahzad Kamal\*



Cite This: *ACS Omega* 2024, 9, 19620–19626



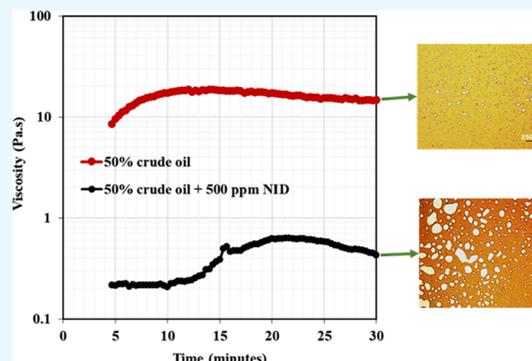
Read Online

ACCESS |

Metrics & More

Article Recommendations

**ABSTRACT:** This study describes how varying oil/water contents affect emulsion formation and the impact they have on emulsion droplet size, viscosity, and interfacial behavior. Crude oil (continuous phase) volume fractions of 40, 50, 60, and 70 vol % were probed in the various W/O emulsions formed. Experimental results from optical morphology revealed the emulsion droplets kept reducing as the crude oil fraction kept increasing, while the droplets were nearly unnoticeable in the emulsions derived from 60 and 70% crude oil. The viscosity–shear rate of emulsions produced from 40, 50, and 60 vol % crude oil exhibited a non-Newtonian behavior owing to the substantial volume of water content in their emulsions, whereas the viscosity–shear rate of the emulsion with 70 vol % crude oil exhibited a Newtonian behavior similar to the pure crude oil, suggesting a thorough blending of oil–water at this crude oil fraction. Besides, the viscosity–temperature measurements revealed that the viscosity of these emulsions diminished as the temperature increased and the viscosity reduction became more noticeable in an emulsion comprising 70 vol % crude oil. In the interfacial assessment, the increased crude oil content in the produced emulsion led to a sharp reduction in the interfacial tension (IFT). The IFT values after 500 s contacts between the emulsion and water (surrounding phase) were 11.86, 10.02, 8.08, and 6.99 mN/m for 40, 50, 60, and 70 vol % crude oil, respectively. Demulsification experiments showed that water removal becomes more challenging with a large volume of crude oil and a small water content. Demulsification performances of the lab-grown nonionic demulsifier (NID) after 10 h of demulsification activity at room temperature (25 °C) were 98, 90, 17.5, and 10% for the emulsions formed from 40, 50, 60, and 70 vol % crude oil, respectively, indicating that the demulsification degree decreases with an increasing crude oil content. Viscosity–time determination was applied to affirm the activity of NID on the emulsion formulated with a 50% crude oil fraction. The injection of NID in this emulsion triggered a sharp viscosity reduction, indicating the adsorption of NID at the oil–water interface and disruption of emulsifiers, enabling emulsion stability.



## 1. INTRODUCTION

Fossil fuel emulsions are invariably encountered in the process of hydrocarbon exploitation, and these emulsions could be in the form of water-in-oil (W/O), oil-in-water (O/W), or multiple emulsions.<sup>1</sup> These emulsions have significant implications for operational design and production in oilfields. Active substances like asphaltenes, resins, and waxes in crude oil have a tendency to form clustered aggregates at the oil–water interface, leading to the formation of a stable emulsion.<sup>1–6</sup> The increasing concentration of these natural active ingredients at the oil–water interface facilitates emulsion formation and further promotes stability;<sup>6</sup> emulsion formation can lead to a significant viscosity change, thereby affecting crude oil production. The majority of natural emulsifying agents in crude oil possess lipophilic and hydrophobic<sup>7–13</sup> characteristics, enabling these natural substances to settle at the

oil–water interface and form rigid interfacial films, preventing coalescence of emulsion droplets. For instance, the viscosity of the W/O emulsion is much related to the oil and water contents and the droplet sizes of the W/O emulsion. As the water content increases, the viscosity of the W/O emulsion tends to increase and the water droplet sizes become more uniform.<sup>6</sup>

**Received:** February 13, 2024

**Revised:** April 3, 2024

**Accepted:** April 11, 2024

**Published:** April 22, 2024



It is well-known that the crude oil production tends to diminish as the reservoir life decreases, causing the water volume to increase. Because of this, fluids produced during the enhanced oil recovery operation are mixtures of water, crude oil, silt, and sand, including organic and inorganic compounds.<sup>13</sup> These mixtures are usually recovered as a crude oil emulsion. Remarkably, researchers have established the fact that the production of emulsions is associated with many challenges such as pipeline and equipment corrosion, pumping cost increases due to the high viscosity of emulsions, and catalyst poisoning. Due to the complex nature of oil reservoirs, it is imperative to gain further understanding on how varying oil and water contents impact the formation of crude oil emulsion and the effects of interfacial adsorption of natural emulsifiers, especially asphaltenes, on emulsion formation, which ultimately affects the production of crude oil in oilfields. In a study carried out by Liu et al.,<sup>14</sup> the inhibiting impact of the composite formulation of kinetic and thermodynamic inhibitors on gas hydrate formation in high water cut oil–water emulsions was investigated so as to deduce how to decrease the operational cost of oil and gas pipelines when confronted with high water content in oilfields. Al-Sakkaf and Onaizi<sup>15</sup> also assessed the effect of acidity, basicity, and salinity on crude oil/water nanoemulsions stabilized by a rhamnolipid biosurfactant. The outcome of their investigation showed that the viscosity of the produced nanoemulsion increased with the increasing pH and salinity level; however, the viscosity remained below 4.2 mPa·s with respect to the crude oil viscosity, and likewise, increasing the temperature of the formulated nanoemulsion led to a viscosity reduction. Furthermore, Yacine et al.<sup>16</sup> worked on the understanding of the rheological and microstructural properties of water/oil and oil/water emulsions. The results of their study showed that the dynamic analysis of emulsions is dependent on the temperature and percentages of water added to the crude oil.

The goal of the current study is to elucidate the effect of oil and water volume ratios on crude emulsion formation and the role they play in the viscosity behavior of an emulsion. Likewise, one of the salient features of this work is to unravel how different oil and water contents affect the interfacial characteristics of the produced emulsion. The emulsifying substances at the oil–water interface travel and adsorb differently when oil and water volumes differ. Hence, interfacial tension measurements of the crude oil emulsion at various oil and water fractions would shed light on how natural emulsifiers such as asphaltenes and resins migrate to the fluid interface. Also, this work reports the demulsification of emulsions formed using a newly laboratory-grown nonionic demulsifier (NID) by condensation of fatty acid with amine. As far as NID is concerned, its use is still limited to demulsification purposes; only a recent study has actually examined the breaking mechanism of water-in-heavy oil emulsions driven by oxygen-enriched NID.<sup>17</sup> Hence, the present investigation carried out further measurements on viscosity and elastic modulus to affirm the effectiveness of NID in the presence of the produced emulsions. This work would definitely be of great interest to oilfield operators as emulsion formation has an enormous impact on crude oil productivity and operational cost.

## 2. EXPERIMENTAL SECTION

**2.1. Materials.** The crude oil used in this study was supplied by oilfield operators, and it was applied directly without any further treatment. Its physiochemical properties

are shown in Table 1. The seawater was formulated in the laboratory using the following salts: Na<sub>2</sub>SO<sub>4</sub>, NaCl, MgCl<sub>2</sub>,

**Table 1. Physiochemical Properties of Crude Oil**

physical characterization and mass percentage	value
API gravity @ 15 °C	32.490
API specific gravity @ 15 °C	0.863
density (gcm <sup>-3</sup> ) @ 15 °C	0.8620
viscosity (mPa·s) @ 15 °C	10.9
SARA fractions:	
saturates (wt %)	36.2
asphaltenes (wt %)	2.8
resins (wt %)	11
aromatics (wt %)	50

NaHCO<sub>3</sub>, and CaCl<sub>2</sub>, with a total salinity of 57,700 mg/L. The precursor chemicals and reagents used in the synthesis of the nonionic demulsifier (NID) were purchased from the Sigma-Aldrich company (Table 2)

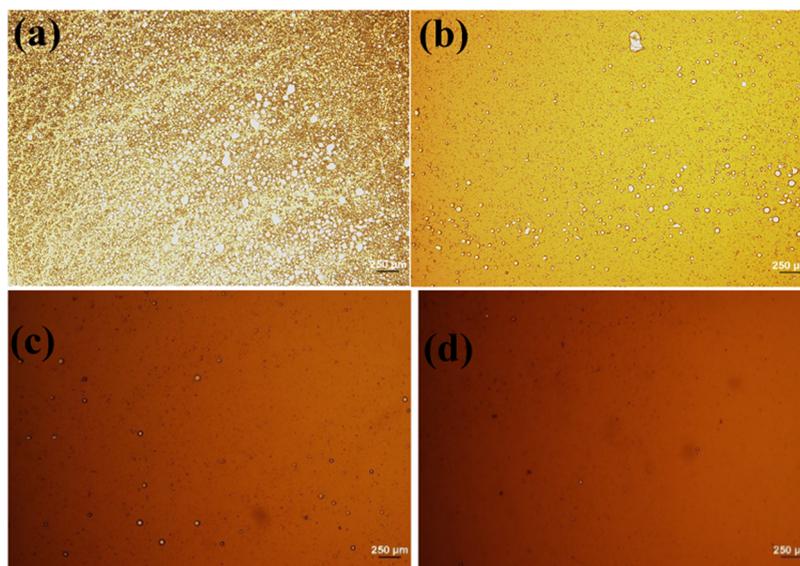
**Table 2. Ions in the Laboratory-Synthesized Seawater**

ions	seawater (mg/L)
Na <sup>+</sup>	18,300
Ca <sup>2+</sup>	700
Mg <sup>2+</sup>	2100
SO <sub>4</sub> <sup>2-</sup>	4300
Cl <sup>-</sup>	32,200
HCO <sub>3</sub> <sup>-</sup>	100
total	57,700

**2.2. Emulsification and Demulsification Methodology.** Crude oil emulsions with different oil/water ratios were formulated. In this study, four emulsions were produced from varying crude oil volume (continuous phase) fractions of 40, 50, 60, and 70%, while the remaining percentage in each emulsion accounts for the water volume. For each emulsion preparation, seawater was added in a stepwise manner inside a 100 mL beaker containing the crude oil, and both fluids were agitated at a speed of 1100 rpm while the mixing was continued for every emulsion formulation until 1 h. It is important to mention that the asphaltenes and resins in the crude oil were utilized directly as emulsifiers and the set of emulsions produced was made to undergo different characterizations before being demulsified by the lab-grown nonionic demulsifier (NID).

The bottle test technique was employed to assess how the NID breaks these emulsions. A 6 g amount of NID was dissolved in a 50 mL bottle containing 14 g of 2-propanol, making the solution 30 wt %. The NID was thereafter diluted to make a 500 ppm concentration that was injected into the produced emulsions. Subsequently, oil and water separation was monitored under the ambient condition (25 °C). All samples were manually stirred for 10 s, and the oil/water separation was monitored periodically. The demulsification efficiency of NID was evaluated using the formula<sup>18</sup> shown in eq 1, and the demulsification rate describing the extent of water removal with respect to time was determined by eq 2.

$$\text{demulsification efficiency (\%)} = \frac{\text{volume of separate water}}{\text{initial water volume in emulsion}} \times 100 \quad (1)$$



**Figure 1.** Optical microscopic images of emulsions having (a) 40%, (b) 50%, (c) 60%, and (d) 70% crude oil as the continuous phase.

$$\text{demulsification rate} = \frac{\text{volume of separated water}}{\text{specific time } (t) \text{ of the separated water}} \quad (2)$$

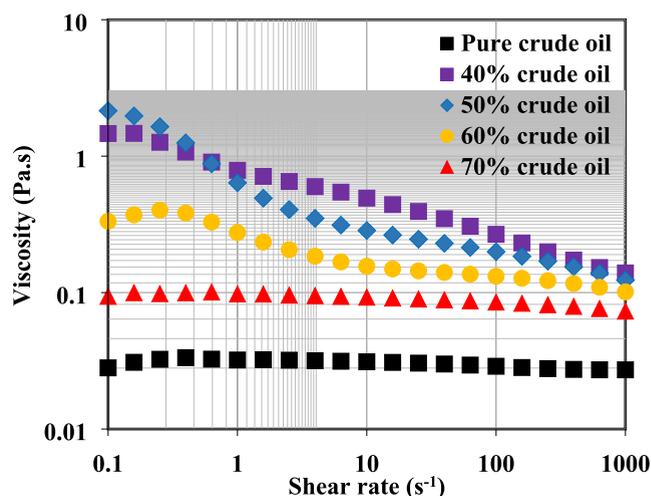
**2.3. Characterizations.** The optical morphology of these emulsions after preparation was analyzed with a high-resolution optical Leica DM2000 microscope. Viscosity and elastic modulus measurements of the produced emulsion before and after the NID injection were conducted at 25 °C using the discovery DHR-3 rheometer manufactured by TA Instruments. The concentric cylinder geometry was chosen with a sample volume of 23 mL for every test. The viscosity–shear rate test was carried out on these emulsions within the range of 0.1–1000  $\text{s}^{-1}$ . During the viscosity–temperature measurements, the shear rate was fixed at 100  $\text{s}^{-1}$ , while elastic modulus tests were carried out at a constant 5% strain, representing the linear viscoelastic region. Interfacial tension (IFT) equipment manufactured by the Biolin Scientific company was employed to gain further insight into the interfacial behavior of the respective emulsion that was released onto the syringe tips inside the seawater as the surrounding phase. In the course of the IFT experiment, around 16  $\mu\text{L}$  of emulsion was systematically released from the capillary tube onto the tip of a J-shaped needle that was dipped inside the seawater. Likewise, the IFT data were recorded for the crude oil released to the syringe tip and dipped inside the NID solution. All IFT experiments were conducted at 23 °C and repeated twice to establish the reproducibility.

### 3. RESULTS AND DISCUSSION

**3.1. Morphology of the Formed Emulsions.** Figure 1a–d shows the optical morphological images of emulsions formed from 40, 50, 60, and 70% crude oil as the continuous phase. It is evident from these images that the formed emulsion from the respective crude oil volume fractions was the W/O emulsion type, and the morphology revealed clearly that the water droplets decreased and became much smaller as the fraction of crude oil increased. The emulsion droplet size became extremely small at 60 and 70 vol % crude oil, suggesting that decreasing the water content in the W/O

emulsion would result in droplets becoming small in size and nearly becoming finely blended with the crude oil. Basically, the average droplet sizes of the W/O emulsions from 40 and 50 vol % crude oil were in the ranges of 2.3–4.5 and 0.9–1.5  $\mu\text{m}$ , respectively, while the droplet size of emulsions with 60 and 70 vol % crude oil was in the range of 0.5–0.9  $\mu\text{m}$ .

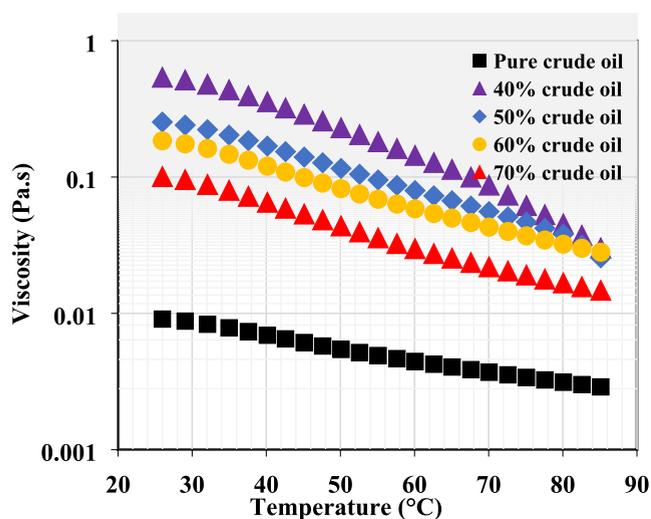
**3.2. Viscosity and Interfacial Behavior of the Formed Emulsions.** It is necessary to stress that the recovery of crude oil in a reservoir becomes extremely challenging because the high viscosity of oil can lead to water fingering<sup>6</sup> and, at the same time, the viscosity of crude oil decreases as the temperature increases. Figure 2 shows the viscosity–shear



**Figure 2.** Viscosity–shear rate profiles of W/O emulsions with varying fractions of crude oil as the continuous phase.

rate profiles of these differing emulsions. As can be observed in these plots, increasing the crude oil content led to a decrease in the emulsion viscosity. Besides, the viscosity of emulsions comprising 40, 50, and 60 vol % crude oil exhibited a non-Newtonian behavior owing to the substantial volume of water in their respective emulsion, whereas the viscosity of emulsion having 70 vol % crude oil demonstrated a Newtonian behavior

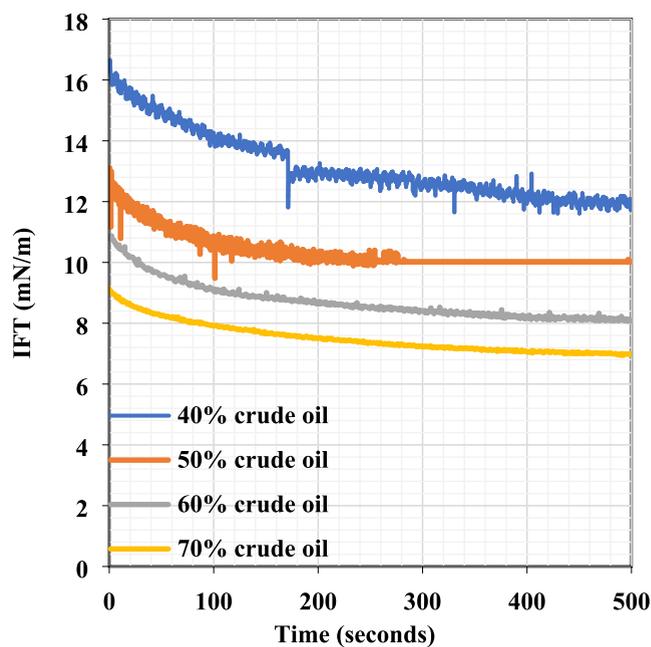
like that of the pure crude oil across the entire shear rate. The Newtonian behavior shown by the emulsion with 70 vol % crude oil revealed the uniform mixing of oil and water and the slip absence between the oil phase and the water phase.<sup>2</sup> At this juncture, it is important to mention that in several instances, the viscosities of W/O emulsions are usually higher than that of the pristine crude oil except in a few cases where a decrease in emulsion viscosity could be observed due to intense shear-thinning and high emulsion instability.<sup>5</sup> It is a known fact that water and a change in temperature would definitely impact the viscosity of crude oil and the W/O emulsion. Because of this reason, the viscosity–temperature profiles of these varying emulsions were studied within the temperature range of 25–85 °C (Figure 3). It is obvious from



**Figure 3.** Viscosity–temperature profiles of W/O emulsions with varying crude oil fractions.

Figure 3 that the viscosity of these emulsions kept decreasing as the temperature kept increasing, and the viscosity reduction was of a high magnitude in the W/O emulsion that was formed from the 70% crude oil fraction. At 85 °C, the viscosities of the W/O emulsion constituting 40, 50, 60, and 70 vol % crude oil were 0.0301, 0.0255, 0.0281, and 0.0148 Pa·s, respectively, as compared to the original (pure) crude oil having a viscosity of 0.0029 Pa·s at this same temperature (85 °C). From the practical point of view in oilfield operations, water injection in a crude oil reservoir can still give rise to emulsion formation at elevated temperatures as the viscosity of the W/O emulsion with 70% crude oil (i.e., having the least water content, 0.0148 Pa·s) was 5.10 times that of the original crude oil at 85 °C. These viscosity–temperature curves supported the fact that emulsification of crude oil and water during oil recovery is practically inevitable and may become more serious as time progresses. Hence, selection of a suitable chemical demulsifier in the downstream process is necessary for effective oil–water separation. Having realized the effect of oil/water content on emulsion viscosity, it is important to address how natural emulsifying agents like asphaltenes affect the interfacial behavior of emulsions formed from different oil/water volume fractions. Several literature studies have actually reported the oil–water interfacial tension properties;<sup>19–22</sup> however, providing insights into the interfacial phenomenon of emulsion/water would be of great relevance in the upstream application. Moreover, examining the interfacial property between the W/

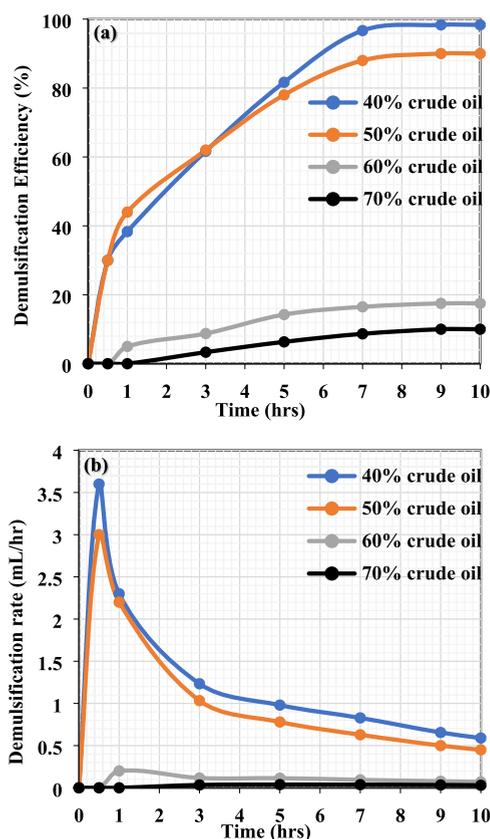
O emulsion and water would provide information on the effect of crude oil content on the produced emulsion. We therefore report the dynamic interfacial tension (IFT) characteristics by making a contact between the individual formulated W/O emulsion–water (as the surrounding phase), and the IFT results are shown in Figure 4. With the increasing crude oil



**Figure 4.** Effect of varying the crude oil content on the dynamic interfacial tension between the produced W/O emulsion and water as the surrounding phase.

content in the formed W/O emulsion, the IFT value was observed to be decreasing. It means that the influx and adsorption of asphaltenes at the interface became higher as the crude oil amount increased in the emulsion. The IFT values after 500 s of contact between the W/O emulsion and water were 11.86, 10.02, 8.08, and 6.99 mN/m for 40, 50, 60, and 70 vol % crude oil, respectively. It is very clear from these findings that varying the degree of crude oil content has an impact on the emulsifier adsorption at the W/O emulsion interface, and at the same time, changing the oil–water content in an emulsion influences the movement and aggregation of asphaltenes at the interface.

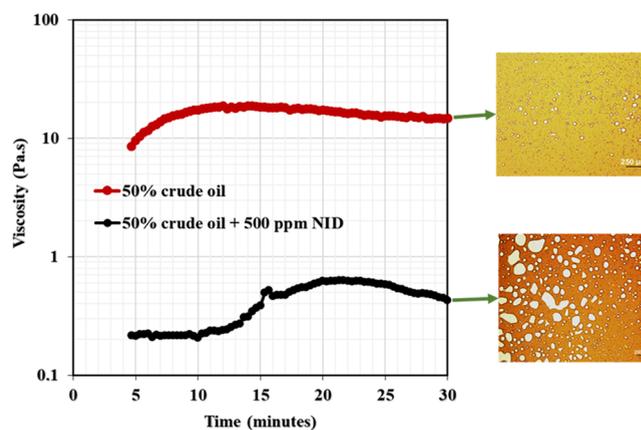
**3.3. Demulsification Induced by a Nonionic Demulsifier (NID).** Irrespective of the crude oil volume fraction, the formulated W/O emulsions showed good stability for more than 30 days by naked-eye visualization at room temperature without any observable oil/water separation. If these emulsions are left for a very long time without being perturbed (without demulsifier addition), the onset of oil and water separation for each emulsion would differ owing to the variation in their droplet size as discussed earlier in Section 3.1. Although an emulsion may have a few advantages in petroleum operation, the emulsion produced during hydrocarbon exploitation could have devastating effects not only on crude oil recovery but also on equipment and operational facilities. It therefore becomes imperative to treat the produced emulsion such that oil and water are separated and disposed appropriately. Figure 5a shows the demulsification performance of a lab-grown nonionic demulsifier (NID) in breaking the W/O emulsions



**Figure 5.** (a) Demulsification efficiency of NID in destabilizing emulsions with varying crude oil contents. (b) Demulsification rate of NID in emulsions with varying crude oil contents. Demulsification was monitored at room temperature (25 °C).

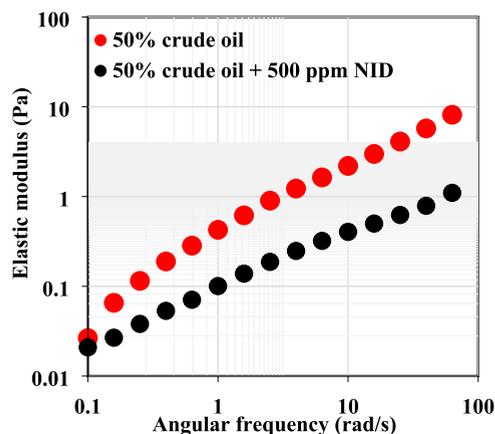
derived from different crude oil fractions. This NID demonstrated excellent demulsifying effectiveness and was able to displace the emulsifiers, enabling the stability of these emulsions. As reflected in Figure 5a, the demulsification efficiencies of NID after 10 h of demulsification activity at room temperature (25 °C) were 98, 90, 17.5, and 10% for the emulsions formed from 40, 50, 60, and 70 vol % crude oil, respectively. It is evident from this finding that the oil–water separation efficiency of NID decreased as the crude oil fraction increased in the emulsion, indicating that water removal became more challenging in the emulsion having 60 and 70 vol % crude oil fractions. As already discussed in the optical morphological analysis of both emulsions in Figure 1c,d, water droplets were very tiny and blended well with the crude oil, making it difficult to extract a substantial volume of water in emulsions derived from 60 and 70 vol % crude oil fractions. Figure 5b illustrates the demulsification rate, highlighting the extent or the speed at which NID was able to remove water in these emulsions. It is very clear that the rate at which NID triggers oil–water separation in emulsions obtained from 40 and 50 vol % crude oil fractions was very fast from the onset until 3 h of demulsification duration; afterward, the demulsification rate of NID declined. In the case of emulsions from 60 and 70 vol % crude oil fractions, the NID demulsification rate was very slow and remained nearly unchanged throughout the 10 h of demulsification activity. In order to affirm the workability of the NID, viscosity–time measurements were performed on the emulsion formulated from the 50% crude oil fraction with and without NID, as

shown in Figure 6. The incorporation of 500 ppm NID in this emulsion resulted in viscosity reduction as compared to the



**Figure 6.** Viscosity–time profiles of an emulsion (50% crude oil fraction) with and without NID. Measurements were conducted at room temperature (25 °C) and 0.01 s<sup>-1</sup> shear rate.

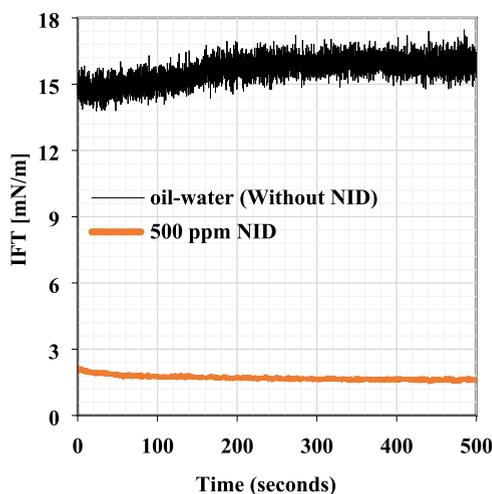
viscosity of the same emulsion without NID. This behavior indicated the aggregation and adsorption of NID at the oil–water interface followed by the disruption of natural emulsifiers, especially asphaltenes.<sup>23–25</sup> The optical morphological images in Figure 6 exhibit the droplet size after the viscosity measurements. The droplet size of the emulsion without NID did not undergo much transformation as compared to the droplets of the emulsion filled with NID. The water droplets of this emulsion changed and increased in size due to the presence of NID, which caused the aggregation and coalescence of the water droplets. In addition, elastic modulus tests shown in Figure 7 revealed that the elasticity of



**Figure 7.** Elastic modulus profiles of an emulsion (50% crude oil fraction) with and without NID. Measurements were conducted at room temperature (25 °C).

the 50% crude oil emulsion filled with 500 ppm NID diminished across the entire frequency region as compared to the same emulsion without NID, indicating that the presence of NID triggered the displacement of elastic interfacial films, enabling the attachment of oil–water molecules at the interface. Interfacial tension measurements were also applied to establish how NID behaves at the oil–water interface. Figure 8 shows the effect of NID on the oil–water interface. It is clear that the presence of NID at the oil–

water interface led to the reduction in the IFT, indicating its adsorption.



**Figure 8.** Dynamic IFT results of the oil–water system with and without NID.

#### 4. CONCLUSIONS

The salient objective of this work is to unravel how varying oil and water ratios impact the morphology, viscosity, and interfacial behavior of an emulsion while using a newly developed nonionic demulsifier (NID) to bring about oil/water separation of various emulsions developed at room temperature (25 °C). These emulsions were formed from the crude oil of 40, 50, 60, and 70 vol % fractions. Experimental results from the optical morphological analysis showed that the droplet sizes of these emulsions kept decreasing with the increasing crude oil fraction, and the droplets became nearly unnoticeable in emulsions formulated from 60 and 70% crude oil contents. The viscosity measurements of emulsions obtained from 40, 50, and 60% crude oil contents exhibited a non-Newtonian behavior, whereas the viscosity of the 70 vol % crude oil emulsion demonstrated a Newtonian behavior like that of the pure crude oil across the entire shear rate. The viscosity–temperature profiles showed that the viscosity of these emulsions decreased with increasing temperature, and the viscosity reduction was the highest in the emulsion formulated from 70 vol % crude oil. The interfacial tension (IFT) of the emulsion was noticed to be decreasing as the crude oil content was increased. The IFT results between the emulsion and water (as the surrounding phase) were 11.86, 10.02, 8.08, and 6.99 mN/m for 40, 50, 60, and 70 vol % crude oil, respectively. The oil–water separation assessments driven by NID after 10 h of demulsification activity at room temperature (25 °C) were 98, 90, 17.5, and 10% for the emulsions formed from 40, 50, 60, and 70 vol % crude oil, respectively. The viscosity and elastic modulus measurements revealed that the NID effectively displaced the asphaltene aggregates at the oil–water interface.

#### ■ AUTHOR INFORMATION

##### Corresponding Authors

Syed Muhammad Shakil Hussain – Center for Integrative Petroleum Research, College of Petroleum Engineering and Geosciences, King Fahd University of Petroleum and

Minerals, Dhahran 31261, Saudi Arabia; [orcid.org/0000-0002-6806-2326](https://orcid.org/0000-0002-6806-2326); Email: [sms Shakil@kfupm.edu.sa](mailto:sms Shakil@kfupm.edu.sa)

Muhammad Shahzad Kamal – Center for Integrative Petroleum Research, College of Petroleum Engineering and Geosciences, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia; [orcid.org/0000-0003-2359-836X](https://orcid.org/0000-0003-2359-836X); Email: [shahzadmalik@kfupm.edu.sa](mailto:shahzadmalik@kfupm.edu.sa)

#### Authors

Ahmad A. Adewunmi – Center for Integrative Petroleum Research, College of Petroleum Engineering and Geosciences, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia; [orcid.org/0000-0002-1673-9649](https://orcid.org/0000-0002-1673-9649)

Shirish Patil – Department of Petroleum Engineering, College of Petroleum Engineering and Geosciences, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia; [orcid.org/0000-0002-0131-4912](https://orcid.org/0000-0002-0131-4912)

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acsomega.4c01400>

#### Notes

The authors declare no competing financial interest.

#### ■ ACKNOWLEDGMENTS

The authors acknowledge the Center for Integrative Petroleum Research at KFUPM for supporting the ongoing research work.

#### ■ REFERENCES

- (1) Umar, A. A.; Saaid, I. B. M.; Sulaimon, A. A. *Rheological and Stability Study of Water-in-Crude Oil Emulsions*, AIP Conference Proceedings; AIP, 2016.
- (2) Li, C.; Huang, Q.; Ma, S.; Ji, C. An Experimental Study on the Viscosity of Water-in-Oil Emulsions. *J. Dispersion Sci. Technol.* **2016**, *37* (3), 305–316.
- (3) Shi, S.; Wang, Y.; Liu, Y.; Wang, L. A New Method for Calculating the Viscosity of W/O and O/W Emulsion. *J. Pet. Sci. Eng.* **2018**, *171*, 928–937.
- (4) Ariffin, T. S. T.; Yahya, E.; Husin, H. The Rheology of Light Crude Oil and Water-in-Oil-Emulsion. *Procedia Eng.* **2016**, *148*, 1149–1155.
- (5) Pang, S.; Pu, W.; Jiang, F.; Gao, H.; Wang, Y.; Chen, Y.; Wei, P. Effect of Water Content on Features of W/O Emulsion during Water Flooding in Heavy Oil Reservoir: Bulk Properties and Mobility Control Characteristics. *J. Pet. Sci. Eng.* **2021**, *207*, No. 109075, DOI: [10.1016/j.petrol.2021.109075](https://doi.org/10.1016/j.petrol.2021.109075).
- (6) Liu, J.; Zhong, L.; Lyu, C.; Liu, Y.; Zhang, S. W/O Emulsions Generated by Heavy Oil in Porous Medium and Its Effect on Re-Emulsification. *Fuel* **2022**, *310* (2021), No. 122344, DOI: [10.1016/j.fuel.2021.122344](https://doi.org/10.1016/j.fuel.2021.122344).
- (7) Farooq, A.; Shafaghat, H.; Jae, J.; Jung, S.-C.; Park, Y.-K. Enhanced Stability of Bio-Oil and Diesel Fuel Emulsion Using Span 80 and Tween 60 Emulsifiers. *J. Environ. Manage.* **2019**, *231*, 694–700.
- (8) Singh, H.; Bhardwaj, N.; Arya, S. K.; Khatri, M. Environmental Impacts of Oil Spills and Their Remediation by Magnetic Nanomaterials. *Environ. Nanotechnol. Monit. Manage.* **2020**, *14*, No. 100305, DOI: [10.1016/j.enmm.2020.100305](https://doi.org/10.1016/j.enmm.2020.100305).
- (9) He, L.; Lin, F.; Li, X.; Sui, H.; Xu, Z. Interfacial Sciences in Unconventional Petroleum Production: From Fundamentals to Applications. *Chem. Soc. Rev.* **2015**, *44* (15), 5446–5494.
- (10) Zafeiri, I.; Horridge, C.; Tripodi, E.; Spyropoulos, F. Emulsions Co-Stabilised by Edible Pickering Particles and Surfactants: The Effect of HLB Value. *Colloid Interface Sci. Commun.* **2017**, *17*, 5–9.

- (11) Raya, S. A.; Saaid, I. M.; Ahmed, A. A.; Umar, A. A. A Critical Review of Development and Demulsification Mechanisms of Crude Oil Emulsion in the Petroleum Industry. *J. Pet. Explor. Prod. Technol.* **2020**, *10*, No. 0123456789, DOI: 10.1007/s13202-020-00830-7.
- (12) Barroso-Solares, S.; Zahedi, M. G.; Pinto, J.; Nanni, G.; Fragouli, D.; Athanassiou, A. Oil Removal from Water-Oil Emulsions Using Magnetic Nanocomposite Fibrous Mats. *RSC Adv.* **2016**, *6* (75), 71100–71107.
- (13) Yonguep, E.; Kapiamba, K. F.; Kabamba, K. J.; Chowdhury, M. Formation, Stabilization and Chemical Demulsification of Crude Oil-in-Water Emulsions: A Review. *Pet. Res.* **2022**, *7*, 459–472.
- (14) Liu, Y.; Duan, J.; Xu, D.; Chu, J.; Shen, S.; Zhao, J.; Jiang, L. Inhibiting Effect of a Composite Formulation of Kinetic and Thermodynamic Inhibitors for Gas Hydrate Formation in High Water Cut Oil-Water Emulsion. *Energy Sci. Eng.* **2023**, *11* (7), 2302–2313, DOI: 10.1002/ese3.1453.
- (15) Al-Sakkaf, M. K.; Onaizi, S. A. Crude Oil/Water Nano-emulsions Stabilized by Rhamnolipid Biosurfactant: Effects of Acidity/Basicity and Salinity on Emulsion Characteristics, Stability, and Demulsification. *Fuel* **2023**, *344*, No. 128052, DOI: 10.1016/j.fuel.2023.128052.
- (16) Yacine, C.; Safri, A.; Djemiat, D. E.; Benmounah, A.; Boumediene, H.; Ezzouar, B.; Alger, A. Rheological Behavior and Microstructural Properties of Crude Oil and Emulsions (Water/Oil/Water). *Pet. Sci. Technol.* **2024**, *42* (9), 1047–1063, DOI: 10.1080/10916466.2023.2232397.
- (17) Zhang, X.; He, C.; Zhou, J.; Tian, Y.; He, L.; Sui, H.; Li, X. Demulsification of Water-in-Heavy Oil Emulsions by Oxygen-Enriched Non-Ionic Demulsifier: Synthesis, Characterization and Mechanisms. *Fuel* **2023**, *338*, No. 127274.
- (18) Adewunmi, A. A.; Kamal, M. S.; Solling, T. I.; Salami, B. A. Palm Oil Fuel Ash (POFA) as a Demulsifier for Crude Oil Emulsions: Performance and Mechanism. *J. Pet. Sci. Eng.* **2019**, *183*, No. 106430, DOI: 10.1016/j.petrol.2019.106430.
- (19) Alves, C. A.; Yanes, J. F. R.; Feitosa, F. X.; de Sant'Ana, H. B. Influence of Asphaltenes and Resins on Water/Model Oil Interfacial Tension and Emulsion Behavior: Comparison of Extracted Fractions from Crude Oils with Different Asphaltene Stability. *J. Pet. Sci. Eng.* **2022**, *208*, No. 109268, DOI: 10.1016/j.petrol.2021.109268.
- (20) Doğan, M.; Saraç, M. G.; Türker, D. A. Effect of Salt on the Inter-Relationship between the Morphological, Emulsifying and Interfacial Rheological Properties of O/W Emulsions at Oil/Water Interface. *J. Food Eng.* **2020**, *275*, No. 109871, DOI: 10.1016/j.jfoodeng.2019.109871.
- (21) Adewunmi, A. A.; Kamal, M. S.; Amao, A. O.; Solling, T. I. Extracted Quartz as Efficient Natural Demulsifier for Crude Oil-Water Emulsions: Effect of Monovalent/Divalent Salts, PH and Modeling Study. *J. Pet. Sci. Eng.* **2021**, *206*, No. 109069, DOI: 10.1016/j.petrol.2021.109069.
- (22) Ahmadi, S.; Khormali, A.; Khoutoriansky, F. M. Optimization of the Demulsification of Water-in-Heavy Crude Oil Emulsions Using Response Surface Methodology. *Fuel* **2022**, *323*, No. 124270, DOI: 10.1016/j.fuel.2022.124270.
- (23) Abdullah, M. M. S.; Al-Lohedan, H. A. Alginate-Based Poly Ionic Liquids for the Efficient Demulsification of Water in Heavy Crude Oil Emulsions. *Fuel* **2022**, *320*, No. 123949, DOI: 10.1016/j.fuel.2022.123949.
- (24) Deng, J.; Zhu, M.; Wang, Z.; Liu, Y. Novel Iron-Modified Chitosan for Effective Demulsification and Oil Recovery of Emulsified Oily Wastewater. *Mater. Chem. Phys.* **2023**, *305*, No. 127848.
- (25) Atta, A. M.; Al-Lohedan, H. A.; Ezzat, A. O. Synthesis and Application of Geminal Dicationic Ionic Liquids and Poly (Ionic Liquids) Combined Imidazolium and Pyridinium Cations as Demulsifiers for Petroleum Crude Oil Saline Water Emulsions. *J. Mol. Liq.* **2021**, *325*, No. 115264, DOI: 10.1016/j.mol-liq.2020.115264.