The role of Gemini surfactant and SiO2/SnO/Ni2O3 nanoparticles as flow improver of Malaysian crude oil

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Original article

The role of Gemini surfactant and $SiO_2/SnO/Ni_2O_3$ nanoparticles as flow improver of Malaysian crude oil

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ABSTRACT

The addition of surfactants and nanoparticles in minimizing wax deposition related problems such as flow assurance is considered an attractive alternative among other techniques but limited researches have been carried out in investigating the method. High viscosity crude oil needs to be treated with viscosity reducer to facilitate transportation and processing. In this study, the efficiency of the viscosity reducer, a Gemini surfactant 2,5,8,11 Tetramethyl 6 dodecyn-5,8 Diol Ethoxylate, three different nanoparticles: silicon dioxide (SiO_2), tin oxide (SnO) and nickel (III) oxide (Ni_2O_3) and their novel blends at different range of concentration, temperature, shear rate and surfactant/nanoparticle loading ratio are assessed in order to study their influence on the viscosity of Malaysian crude oil using Brookfield DV-III viscometer. The separate use of Gemini surfactant and nanoparticles alone resulted in significant reduction in crude oil viscosity. The combined use of Gemini surfactant and nanoparticles showed better performance as compared to their corresponding individual use. The presence of surfactant improves the adsorption of nanoparticles by functionalizing their surfaces. Adsorption and adhesion of wax molecules onto the surface of nanoparticles and surfactants prevents them from aggregating. This results in increment in viscosity reduction. Overall, the viscosity of crude oil was reduced about 85–92% at 10 °C with the aid of Gemini surfactant and nanoparticles. The highest viscosity reduction obtained was 92.8% using the blend of Gemini surfactant and silicon dioxide. The findings of the study are expected to contribute to the crude oil industry in improving flow in production and transportation.

1. Introduction

As a form of fossil fuel, crude oil can be refined to produce usable goods such as petrol, diesel and similar petrochemical forms. A major issue faced by the crude oil industry is the deposition of paraffin wax from low-temperature crude oils, which may block pipelines. During transportation, the wax particles in crude oil precipitate and deposit on the pipe wall in subsea region due to temperature gradient. The temperature of the crude oil transportation pipeline is lesser than 10 °C and can reach 4 °C due to cold environment (Ahn et al., 2005; Akinyemi et al., 2018; Lim et al., 2018; Lucas and Machado, 1999; Ridzuan et al., 2015). Paraffins can crystallize from crude oil and increase the oil's viscosity, thereby impeding the flow and blocking the pipeline.

The use of chemicals to boost the flow of crude oil at low temperatures is a readily accessible and well-recognized solution that is economically feasible for pipeline blockage issue. Machado et al. (2001) and Anisuzzaman et al. (2017) used Poly(ethylene-co-vinyl acetate) (EVA) to reduce viscosity of crude oil. EVA is also one of the frequently used polymers to inhibit wax deposition, and their effects on pour point, cloud point and performance inhibition are often investigated. Whereas, Subramanie et al. (2019) investigated EVA with the incorporation of silica nanoparticle and compared it with Poly (maleic anhydride-alt-1-octadecene) (MA) on the performance as viscosity reducer. The research found that the individual usage of EVA and MA achieved viscosity reductions by 88% and 86.4%, respectively while when they were added with nanoparticle, about 94% and 89.2% viscosity reduction were achieved, respectively. The addition of nanoparticle resulted in greater efficiency.

Few researches investigated the use of nanoparticles in viscosity reduction of crude oil such as magnetite (Fe_3O_4) nanoparticle-based ferrofluids (Aristizábal-Fontal et al., 2018), metal oxide

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nanoparticles (CuO, NiO and Fe_2O_3) (Patel et al., 2018) and iron oxide nanorods (Al-Ruqeishi et al., 2019). Patel et al. (2018) found that each type of nanoparticle used achieved 50–70% viscosity reduction and the findings showed that there was an optimal concentration of nanoparticles that achieved the highest decrease in viscosity. Nanoparticles exhibit unique material characteristics such as high surface area due to their particle size and have practical applications in different industries. Also, due to the cost-effectiveness and environmentally friendly approach, nanotechnology is more attractive.

Nanoparticles tend to aggregate because of high surface area and surface activity. A significant technique to improve the stability of nanoparticles is by using surfactants. A number of researchers found that the blend of nanoparticles and surfactants has better performance than their individual use. Lim et al. (2019) investigated the performance of silane-based surfactant (SN3) and silicon dioxide nanoparticles (SiO_2 NPs) on the viscosity reduction of light crude oil. The crude oil viscosity was effectively reduced by SN3 alone by 51% while its blend with SiO_2 NPs showed the highest DVR (67%). Whereas, in Lim et al. (2018)'s research, the overall finding found that the addition of SiO_2 nanofluid into surfactant-treated crude oil gave better PIE performance, as compared to the surfactants without nanofluids.

In terms of surface activity, Gemini surfactants are generally superior over a conventional surfactant (Sekhon, 2004). Although Gemini surfactants are widely used in enhanced oil recovery (EOR) (Hou et al., 2019; Pal et al., 2018a, 2018b), limited studies have been carried out to research theirs potentials and applications in reducing viscosity of crude oil. However, studies have been carried out on the application of Gemini surfactant for wax dispersants by Maithufi et al. (2011) which reduced the amount of crystals and sizes. Also, Sahai et al. (2018) investigated laboratory synthesized Gemini surfactants to reduce viscosity of bituminous crude oil and found that, they improve the flowability by restraining the self-aggregation of asphaltenes.

In this work, three different nanoparticles and a Gemini surfactant as viscosity reducer separately, and the blends were evaluated individually as viscosity reducers of Malaysian light crude oil. As far as the authors are aware, to date, there is no published analysis on the performance of Gemini surfactant 2,5,8,11 Tetramethyl 6 dodecyn-5,8 Diol Ethoxylate with silicon dioxide, tin oxide and nickel (III) oxide and their blends as viscosity reducer of Malaysian crude oil. These types of nanoparticle in reducing the viscosity of Malaysian crude oil has not been extensively studied. Therefore, it is sensible to study the efficiency of the Gemini surfactant and nanoparticles in order to realize their maximum potential in mitigating flow assurance problem. The present study is an effort to evaluate the influence of viscosity reducer and nanoparticles at different concentrations, surfactant/nanoparticle blending ratios, temperature and shear rate on the viscosity.

2. Methodology

2.1. Materials

The light crude oil sample was provided by PETRONAS Penapisan Terengganu Sdn. Bhd. and the properties are shown in Table 1. Gemini surfactant (GS) 2,5,8,11 Tetramethyl 6 dodecyn-5,8 Diol Ethoxylate was used as viscosity reducer which was supplied by Evonik Corporation and the molecular structure is shown in Fig. 1. The nanoparticles used were silicon dioxide (SiO_2:NP1), tin oxide (SnO:NP2) and nickel (III) oxide (Ni_2O_3:NP3) purchased from Nanou Nanotechnology with particle sizes of 20 nm, 20 nm and 50 nm, respectively.

Table 1
Properties of Malaysian crude oil (Subramanie et al., 2019).

Physical properties	Value
WAT, °C	28
Pour point, °C	11
Wax Content at 20 °C, %	9.2
Density, g/cm^3	0.814
°API	42.4
Chemical Properties	Percentage %
Saturates	40.83
Aromatics	32.08
Resin	11.67
Asphaltene	15.42

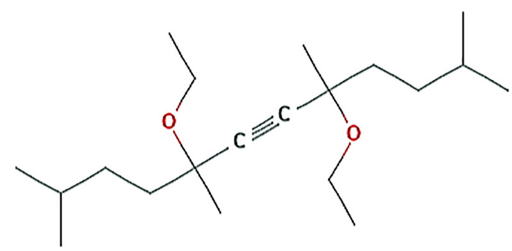

Fig. 1. Molecular structure of 2,5,8,11 Tetramethyl 6 dodecyn-5,8 Diol Ethoxylate.

2.2. Preparation of nanofluid

To prepare 100 mL of SiO_2 solution with 200 ppm concentration, 0.02 g of SiO_2 powder was mixed with 100 mL of cyclohexane and stirred for 2 hrs at 60 °C and 700 rpm followed by ultrasonication treatment for a time period of 1 hr at 25 °C. SiO_2 solutions of 300 ppm, 400 ppm, 500 ppm and 600 ppm were prepared by dissolving 0.03 g, 0.04 g, 0.05 g and 0.06 g of SiO_2 in cyclohexane of 100 mL, respectively. The preparation procedure was repeated for SnO and Ni_2O_3, which were the other nanofluids used.

2.3. Preparation of GS and nanoparticles at different ratios

Three different ratios of GS to nanoparticles blend were prepared, which were (i) 3:1, (ii) 1:1 and (iii) 1:3 for all types of nanoparticles. 3:1 ratio GS/nanoparticles blend was prepared by mixing 75% of GS and 25% of nanofluid, making up a total of 2 mL, which was added to 7 mL of crude oil, and later analyzed for viscosity and DVR performance. For ratios 1:1 and 1:3, 50% of surfactant with 50% of nanofluid and 25% of surfactant with 75% of nanofluid were mixed, respectively, making up a total volume of 2 mL.

2.4. Characterization of nanoparticles dispersion

UV–vis absorption spectra were recorded with a Thermo Fisher Scientific spectrometer. UV–Vis spectral analysis is an accurate method of analysis of dispersion stability of nanofluids. UV–vis spectral analysis is a reliable and accurate approach to determine nanofluids' stability if the nanoparticles distributed in fluids possess characteristic absorption bands within the wavelength 190–1100 nm (Yu and Xie, 2012). Cyclohexane is used as reference standard and nanofluids as test samples to evaluate the dispersion, and the absorbance values were estimated at a region between 400 nm and 800 nm.

2.5. Viscosity analysis

The Brookfield Viscometer model DV-III with spindle no 31 was used to measure the crude oil viscosity at various shear rates (80 rpm to 200 rpm). The viscosity measurement was conducted using GS solution with and without the addition of nanofluid. Crude oil sample was heated at 70 ℃ to completely dissolve wax crystals. To conduct the test, a sample vessel of 7 mL of crude oil and 2 mL of GS with and without the addition of nanofluid was prepared. The temperature was manipulated, and controlled using a thermostat circulating bath. The reading indicated in viscometer display was taken after a period of time when stabilization has been achieved, whereby the average value was determined. The temperature was varied within the range of 10 ℃ to 30 ℃. The degree of viscosity reduction (DVR) is applied to determine the magnitude of viscosity reduction and can be determined using the following Equation (1):

$$(\mathrm{DVR})\% = \frac{initial\,value - final\,value}{initial\,value} \times 100 \quad (1)$$

3. Results and discussion

3.1. UV–visible spectroscopy of nanofluids

UV–Vis is applied to measure absorption in liquid samples. The highest possible UV–Vis absorbance also was proposed by several researchers as a good measure of maximum possible dispersion in sonication method (Yu et al., 2007). The length of treatment as well as the energy of the ultrasonication method has a great impact on nanoparticles dispersion. However, within limits of a certain length of time, longer duration of treatment and higher ultrasonic energy, results in better dispersion (Njuguna et al., 2015). Fig. 2 shows the absorbance value for each nanoparticle solution with and without GS. It was found that the absorbance value for nanofluids NP1, NP2 and NP3 are lower than their corresponding blends with GS. Higher value of absorbance indicates better dispersion and less aggregation of nanoparticles.

Surfactants' use in nanofluids also were termed dispersants. Dispersants can noticeably improve the surface properties of a system in low proportions and are employed to enhance the interaction of two materials, which is also described as wettability (Yu and Xie, 2012). The addition of GS to nanofluids has caused the nanoparticles to segregate from each other and has prevented them from forming agglomerates, so that, due to dispersion, a higher surface area of nanoparticles could be achieved. Therefore, greater absorption in UV–Vis spectra indicates better dispersion (Safaei-Naeini et al., 2012).

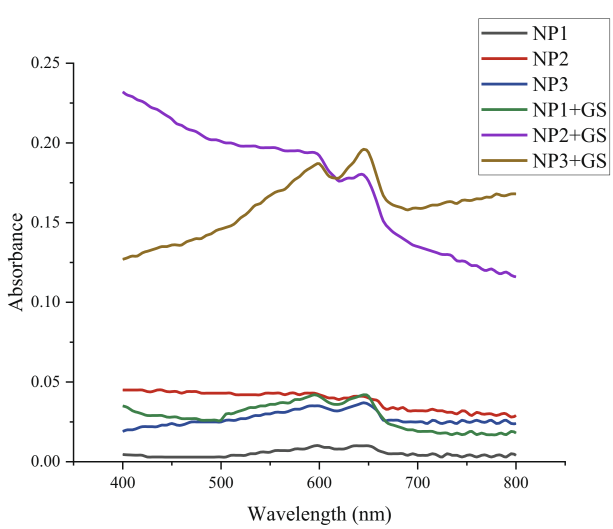

Fig. 2. UV spectrum for nanofluids with and without GS.

3.2. Effect of temperature and shear rate on crude oil viscosity

Two variables affect the crude oil viscosity: temperature and shear rate of crude oil. Fig. 3 represents the effect of shear rate (80 rpm – 200 rpm) and temperature (10 ℃–30 ℃) on crude oil viscosity. According to Hemeida (1990), all fluids that do not exhibit a direct proportionality to shear rate at constant temperature are non-Newtonian fluids. From Fig. 3(a), it can be observed that over the range of shear rates evaluated in which the temperature was kept at a constant at 10℃, both doped and undoped crude oil show non-Newtonian shear thinning behaviour as the viscosity decreases with increasing shear rate. At high shear rates, the energy exerted disrupts the bonds between wax structures and reduces wax formation, resulting in low viscosity (Behbahani et al., 2011).

There was a notable drop in viscosity of both doped and undoped crude oil over the temperatures measured as depicted in Fig. 3(b) at which the shear rate was kept constant at 80 rpm. The viscosity of undoped crude oil dropped from 51.95 cP at 10 ℃ to 3.75 cP at 30 ℃. When temperature decreases below WAT (28 ℃), the amount of dissolved wax begins to exceed its limit of saturation, creating a solid solution in the crude, resulting in a sharp rise in viscosity (Taraneh et al., 2008).

In addition, since cyclohexane was used as solvent for viscosity reducer and nanofluids, its effect on viscosity of crude oil was

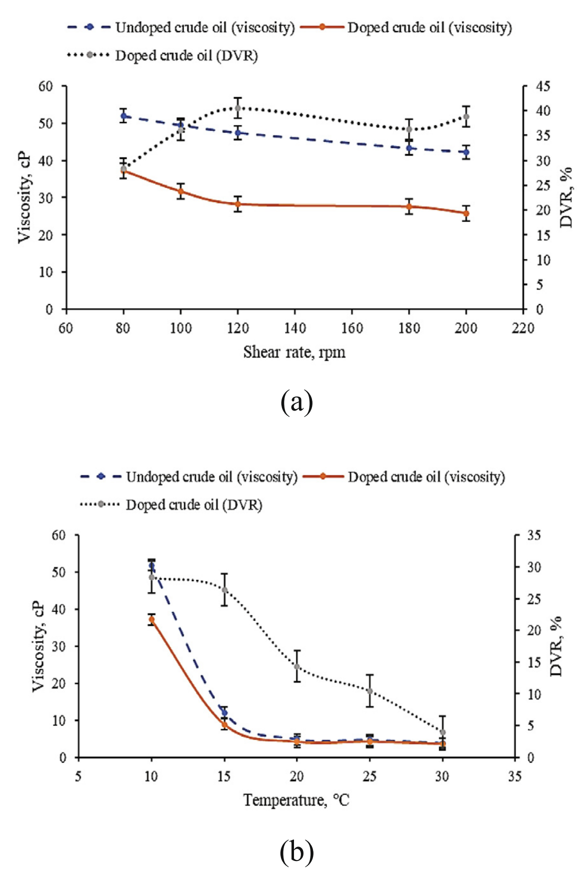

Fig. 3. Effect of (a) shear rate and (b) temperature on the viscosity of undoped and doped (with cyclohexane) crude oil.

investigated. The reduction in viscosity was more significant with the addition of viscosity reducer and nanoparticles compared to cyclohexane. The reduction in viscosity of crude oil is depended on the type and concentration of additives used, as discussed in the following sections.

3.3. Effect of GS and nanoparticles at different concentration on viscosity of crude oil

By plotting DVR as a function of GS and nanoparticle concentration, the best concentration was determined. Fig. 4 and Fig. 5, respectively, show the effect of GS and nanofluids on the viscosity of crude oil at different concentration with constant temperature (10 ℃) and shear rate (80 rpm). The viscosity decreased with increasing concentration of GS until an optimum value was achieved, and then, the value started to decrease slightly. In Fig. 4, it can be seen that, crude oil viscosity decreased from 51.95 cP to 4.5 cP with 400 ppm GS, with the highest degree of DVR at 91.34%. Viscosity of crude oil could not be decreased by raising the concentration to 1000 ppm. Therefore, in this case, the overall concentration of GS cannot exceed 400 ppm to reduce the viscosity of crude oil, whereby 400 ppm is concluded to be the best concentration of GS as viscosity reducer among the tested range of concentration.

Further screening was conducted to find the best concentration of mixture in nanoparticles. The DVR of nanoparticles with and without GS was investigated with concentrations of 200 ppm to 600 ppm. Based on Fig. 5, the best viscosity reducing nanofluid concentrations assisted with GS were 200 ppm, 300 ppm and 400 ppm of NP1, NP3 and NP2, respectively, within the range of the concentrations studied. Further increase in nanofluid concentration does not decrease the viscosity. The reason behind this can be the properties of nanoparticles that serve as extra suspended particles and aggravate the flow of crude oil (Lim et al., 2018). NP1, NP2 and NP3 nanofluids without the addition of GS gave the highest DVR at 600 ppm, 300 ppm and 600 ppm, respectively within the tested ranges of concentration, as depicted in Fig. 5.

The viscosity decreased with increasing concentrations of nanofluids until an optimal concentration is attained. This may be attributable to an increase in the particles' packing factor causing nanoparticles aggregation and reduces the interaction energy among wax aggregates (Taborda et al., 2017). Also, at higher concentrations, nanoparticles cause oxidation of liquid hydrocarbons and form larger molecular size structure resulting in viscosity increment (Jia et al., 2016). This can explain the decrease in viscosity when the concentration of certain nanofluids with and without GS further increased up to 600 ppm.

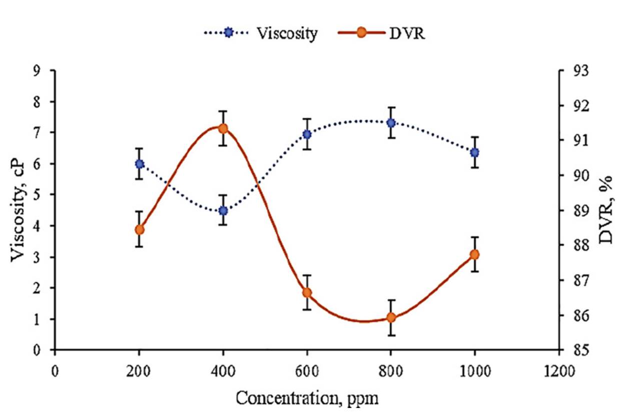

Fig. 4. Viscosity and DVR of crude oil with GS of different concentration.

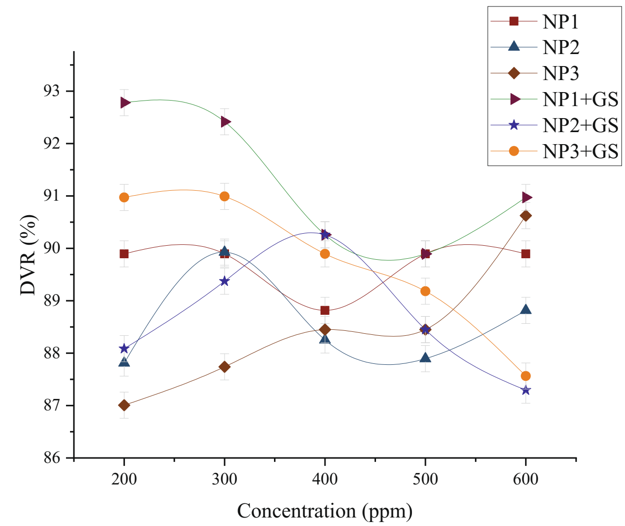

Fig. 5. DVR of different concentrations of nanofluids with and without the assistance of GS (GS concentration was fixed at 400 ppm).

3.4. Effect of GS and nanoparticles at different temperature and shear rate on DVR

Upon the addition of GS and nanoparticles, there was a reduction in crude oil viscosity at all shear rates and temperature. From Fig. 6, GS and nanoparticles are observed to be exhibiting a non-Newtonian behavior, in which the viscosity is influenced by shear rate. The DVR decreased when the shear rate was increased up to 100 rpm, which can be due to a partial internal structure breakdown. Further increase in shear rate resulted in increasing DVR. When the system was subjugated to shear rates higher than 100 rpm, the wax particles appear to disaggregate from their original condition of unstirred mechanism (Taborda et al., 2017). This causes reduction in viscosity. Following that, the increasing shear rate reduced the DVR, which was mostly owing to a change in the fluid's internal structure that caused the viscosity to decrease (Taborda et al., 2017). Reduction in DVR happened due to the behaviour of shear thickening from a very viscous fluid (Ridzuan et al., 2015).

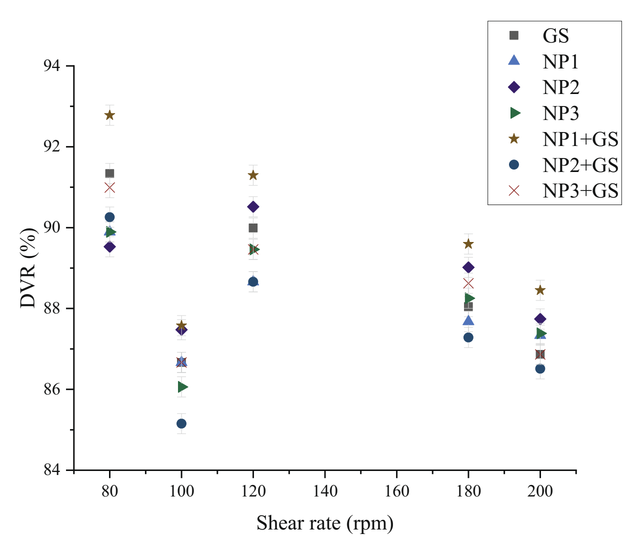

Fig. 6. Degree of viscosity reduction at different shear rates.

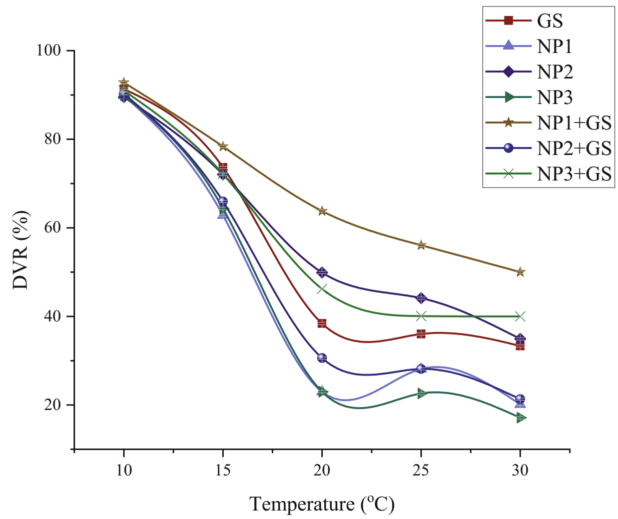

Fig. 7. Degree of viscosity reduction at different temperature.

From the results in Fig. 7, GS and nanoparticles were able to reduce the viscosity of crude oil at all tested range of temperature. The viscosity alteration ranged from 15 to 50% at 30 ℃ to as high as 89–93% at 10 ℃. It was found that at temperatures near the pouring point, the flow improvers decreased the viscosity of the crude oil and the results were dependent on the type of flow enhancer used. Generally, at temperature of 10 to 15 °C, the greatest effect of viscosity reduction can be observed. The crude oil region switched from Newtonian to non-Newtonian fluid region below pour point temperature given the presence of solid particles (Ridzuan et al., 2015). The oil viscosity closer to the WAT was almost unaffected by GS and nanoparticles. This is partially because, in any event, at those temperatures, there was just a small amount of solid wax (Pedersen and Rønningsen, 2003). The possible impact of wax crystal modifiers is therefore, minimal.

3.5. Effect of GS/nanoparticle loading ratio on the DVR

Fig. 8 represents the effect of GS/nanoparticles blend ratio on the DVR at 80 rpm and 10 ℃. For NP1 and NP2, the value of DVR increased in the following order: 3:1 > 1:3 > 1:1 and for NP3, the value increased in the order of 3:1 > 1:1 > 1:3. This is due to surface activity of surfactant molecules that changes resulting from the interaction between nanoparticles and GS. If the ratio of concentration between surfactants and nanoparticles is relatively low, surfactants are capable of covering only a tiny portion of the surface of a nanoparticle. However, a double coating of surfactants on

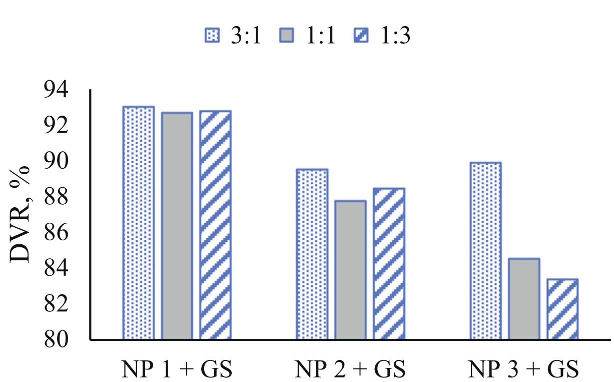

Fig. 8. Addition of nanoparticles to GS at different loading ratio.

the nanoparticles may be formed at larger concentration ratios (Almahfood and Bai, 2018).

The addition of nanoparticles is capable of enhancing the surface activity of surfactants, though only the concentration and load are favorable (Lim et al., 2018). Higher surface activity results in higher reduction in crude oil viscosity. Thus, higher surfactant to nanoparticle ratio is better for higher surface activity. Also, the use of GS altered the flocculation and hydrophobicity of nanoparticles and thus increased the stability. Surfactant serves as a stabilizing agent, restricting the reaggregation of nanoparticles and thus decreasing their size (Paramashivaiah and Rajashekhar, 2016).

3.6. Performance of GS and nanoparticles in viscosity reduction

Fig. 9 shows the extent of viscosity reduction achieved by GS and each nanoparticle with and without GS. NP1 + GS gave the highest viscosity reduction, which is 92.78% of DVR. NP1 + GS gave the highest viscosity reduction, which is 92.78% of DVR. Whereas, NP2 shows the lowest viscosity reduction with a DVR of 89.53%. Nevertheless, the difference is less significant. The order of viscosity reducer and nanoparticle performance in reducing viscosity in descending order is as follow;

NP1 + GS > GS > NP3 + GS > NP2 + GS > NP1 > NP3 > NP2

The performance of GS assisted with nanoparticles were better as compared to the performance of nanoparticles without GS. The findings have shown that the use of GS can change the surface of nanoparticles to contribute to the reduction of viscosity. In light crude oil, the viscosity is caused by agglomeration of wax particles. Due to the existence of elevated affinity for adsorption and wide surface area of nanoparticles, the addition of nanoparticles can facilitate wax molecules dispersion and stabilization of crude oil.

The underlying mechanism is that, surfactant molecules prevent the formation of a three-dimensional interlocking network structure and thus interfering with the process for wax crystal growth (Wang et al., 2016). With reference to Fig. 7, this is also an agreement with the substantial viscosity reduction below WAT, at which wax begins to form. According to Lim et al. (2018), by altering the crystal structure itself, the use of surfactants is capable of modifying wax deposition's layer surface and controlling the rigidity, adhesion or quantity of wax deposits.

Due to their high affinity of adsorption and large surface area, the use of nanoparticles alone helps in dispersing wax molecules. This activates the adsorption of wax particles onto nanoparticles' surface. As a result, the aggregation of wax particles reduces. In addition, in order to functionalize and improve the adsorption of the surface of nanoparticles, the presence of surfactants in the emulsion is also essential (Lim et al., 2018). This explains the rea-

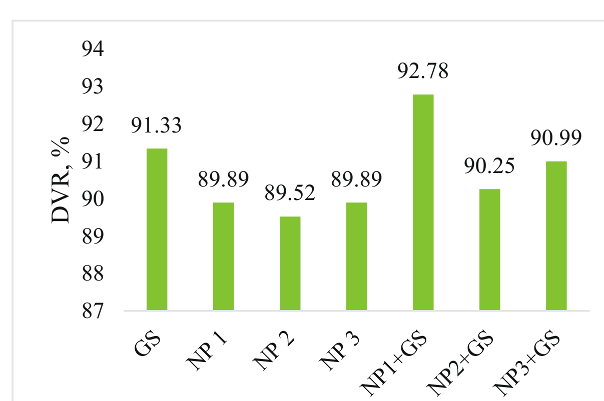

Fig. 9. Degree of viscosity reduction at a fixed shear rate and temperature of viscosity reducer and nanoparticles.

son behind the higher DVR value of NP1 + GS, NP2 + GS and NP3 + GS as compared to their corresponding individuals. Further work is typically needed to elucidate the surfactant/nanoparticle blend mechanism.

Moreover, the adsorption of nanoparticles and surfactant molecules onto the wax molecules stabilizes paraffin emulsion, resulted in viscosity reduction. According to Hao et al. (2019), viscosity reduction occurs due to the co-crystallization process mechanism. This happens when wax inhibitor disrupts the process of crystallization and modifies wax crystal growth. Inhibitors with a similar chemical structure adsorb wax molecules on the surface, which are then bound together and then form a lattice structure of wax crystal in the crude oil. It leads to the modification of growing wax crystals' morphology and disruptions in the formation of three-dimensional crystals.

An effective viscosity reducer, in industrial terms, is capable of forming weaker and softer deposits that are more likely to be worn off by shear forces in the flow area (Frigaard et al., 2017). A surface-active agent's purpose is to provide an effective promotional coating that can counterbalance the attractions of van der Waals to cause electrostatic or steric repulsions (Paramashivaiah and Rajashekhar, 2016). To restate, though the use of nanoparticle on its own can reduce the viscosity of crude oil, GS-assisted nanoparticles showed higher viscosity reduction.

4. Conclusion

By reducing the viscosity, the addition of GS and nanoparticles increases the flow capability of crude oil. However, some parameters, such as concentration, temperature, and shear rate, impact the efficiency of GS and nanoparticles. Moreover, the blends of GS and nanoparticles gave better reduction in viscosity as compared to the use of nanoparticles and GS, respectively, though the difference is not substantial. This can be due to the addition of GS, that modifies the surface of nanoparticles to increase their surface adsorption onto wax molecules. As a consequence, the aggregation of wax molecules will be reduced, which will result in lower viscosity.

The study's other notable conclusion is the influence of temperature and shear rate on the crude oil viscosity. For temperatures near 10° C, a significant reduction in viscosity is observed while the viscosity reduction at higher temperatures is smaller. This study found that when the shear rate increased, the viscosity of crude oil is observed to be decreasing. The findings of the study are expected to contribute to the crude oil industry in improving flow in production and transportation.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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