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Surfactant-solid complex for enhancing the flow in pipelines: an experimental approach

Wafaa K. Mahmood¹, Makarim H. Abdulkarim¹ and Hayder A. Abdulbari^{2*}

Abstract

Background: Viscoelastic soluble polymeric additives have been used successfully for a long time as drag reducers in pipelines carrying commercial liquids like crude oil. Most of these polymers suffer from irreversible degradation when exposed to high shearing zones as in valves, elbows, and pumps which reduces, or eliminates, its flow enhancement effect. Insoluble additives were proven to be an effective drag reducer that overcomes the degradation drawback of soluble additives. On the other hand, insoluble additives suffer from the lack of viscoelasticity which limits their use as flow enhancers. The creation of complexes from soluble and insoluble additives is a field of research that is rarely explored despite its importance in introducing new flow enhancement methods for a higher drag reduction performance. The present work introduces a new surfactant–solid complex as a drag-reducing agent for turbulent flow in pipelines.

Results: The surfactant, solid, and their complexes' drag reduction performance was tested in a closed-loop turbulent flow liquid circulation system, while rheological characteristics of the soluble additives were tested using a standard rheometer. All the surfactant solutions showed non-Newtonian shear thinning behavior in all the investigated concentrations that ranged between 500 and 1300 wppm. The initial experimental result indicated that the surfactant solution's drag reduction performance was higher than that of the solid suspensions. On the other hand, the drag reduction performance was enhanced by 52% when creating a 1300 wppm surfactant–2000 wppm solid complex. This improvement in the drag reduction performance is due to the formation of surfactant–solid-enforced aggregates with high resistance to shear forces and high turbulence suppression efficiency.

Conclusions: The present work introduces a new drag reduction solid–surfactant complex by creating aggregates combining the viscoelastic properties of surfactants with the resistance to high shear forces exerted by the solid particles. The polar nature of the surfactant micelles that form in single-phase flow systems contributed significantly to trapping the solid's micro-particles as enforcement to resist the shearing forces applied by the turbulent flow system.

Keywords: Drag reduction, Turbulence, Surfactants, Sand, Viscoelasticity

1 Background

Enhancing the flow in pipeline liquids and flowing in turbulent flow mode attracted the attention of many researchers around the globe since the early observation by Tom et al. [27] in the early 40s of the past century. Fundamentally speaking, the addition of minute quantities

of soluble viscoelastic additives like polymers and surfactants was proven to have a specific impact on the flow behavior inside the pipelines at turbulent flow mode. Since turbulent flow systems are classified as chaotic with unrepeatable flow behavior, several mechanisms explaining the drag reduction phenomenon were suggested by several authors [9, 14, 23, 29, 31]. These mechanisms, in many cases, contradict due to the explanation philosophy of the researchers. Lumley [15, 16] introduced the molecular extension theory, where coiled polymeric chains are stretched when exposed to the high shear force of the

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turbulent flow systems. Hinch [4] suggested four models that explain the drag reduction theory with polymeric additives: elastic ellipsoid model, inextensible flexible thread model, transversely diffusing thread model, and elastic rod model. De Gennes [26] introduced the elastic modulus to explain the drag reduction phenomenon. They suggested that the polymeric additives viscoelastic characteristic controls the drag reduction behavior where at high turbulence zones, the polymer additive's elastic characteristic will help suppress the eddies forming the turbulence and hence increase the flow. Despite the large number of mechanisms suggested to explain the drag reduction phenomena with soluble additives, especially polymers, these phenomena may be summarized into two major effects. The first phenomenon suggests that the polymeric additives introduce new viscoelastic properties to the eddies and prevent them from forming or completing their shape, and that will result in reducing the number of eddies absorbing energy from the main flow, which will reduce the back-flow phenomena that cause pumping power losses [24, 30]. The other mechanisms suggest that the polar nature of the additives will enable their migration to the pipe's internal walls to neutralize and that will embolize the laminar sublayer, which will result in reducing the flowing liquid layers friction that usually causes the formation of eddies [5, 21]. Either way, high molecular weight polymeric drag reducing agents (DRAs) were experimentally proven to be an effective additive for enhancing the flow in pipelines when introduced in very low concentrations. On the other hand, polymeric DRAs suffer from several drawbacks that minimize their usage and increase their operating costs. The main drawback is the polymeric DRA's resistance to mechanical degradation when exposed to high shear forces in elbows, valves, and pumps. The complex nature of the long-chained polymeric additives will not always provide good resistance when exposed to shearing forces, and any break in their C-H chains will result in an irreversible creation of smaller polymeric molecules, in many cases monomers, that have a much lower drag reduction effect [6]. Several solutions were suggested to overcome this drawback, like restructuring the polymer itself and increasing its resistance to mechanical degradation or reinjecting new polymeric additives after a certain flow distance. It is essential to highlight that both solutions will increase the operation and raw materials costs.

Surface active agents (surfactants) attracted the attention of many researchers to be used as DRAs due to their unique polar characteristics that are believed to overcome the polymeric DRA's mechanical degradation drawback. Surfactants' molecular structures have either polar heads or polar tails in most types and are used as demulsification or emulsification agents when introduced

to a two-phase system [7, 10, 11]. On the other hand, the surfactant molecules in a single-phase system will tend to create aggregates called micelles. These aggregates will act as one large molecule that can partially simulate the polymeric DRAs flow enhancement performance. When these aggregates are exposed to high shearing forces, they tend to break up easily to their initial individual molecules. After passing the high shearing zones, the separated surfactant molecules will reform and create the same micelles again due to their polar nature. Such characteristic is important to overcome the irreversible polymeric DRAs degradation problem, especially when carrying commercial liquids in strategic pipelines.

Insoluble additives are used as flow enhancers to challenge the solubility condition for any additive to be classified as a DRA [13]. It is believed that the interaction of powders within a certain particle diameter (usually less than 600 μm) with the turbulence structures (eddies) will result in suppressing these eddies and reducing the drag [3, 12, 19]. The mechanism controlling this phenomenon is not clear since the additives, in this case, are insoluble, and no modification occurs to the transported liquid physical properties.

The creation of complexes from different soluble additives is one of the approaches to overcome the mentioned drawbacks, especially with polymeric DRAs. Several attempts are spotted in the literature to create different polymer-surfactant complexes as new or modified DRAs that have the viscoelasticity and the structure reinforcement provided by the polar nature of the surfactant's molecules [1, 25]. The polar nature of the surfactant's molecules will allow them to migrate and attach themselves to the polymeric molecules that are oppositely charged. This will create a bonding environment where the presence of the surfactant's molecules on the polymers network surface will enable the bonding of nearby polymeric molecules as well, resulting in larger macroaggregates that act as one unit with higher resistance to mechanical degradation. Mahmood et al. [17] created a polymer-surfactant complex using chitosan polymer and sodium laurel ether sulfate surfactant and used it as a DRA in a water flow system. They found that the drag reduction performance of the proposed complex has a higher resistance to mechanical degradation and higher drag reduction performance compared to the performances of the individual additives. Matras and Kopiczak [18] created a DRA complex from poly(ethylene oxide) polymer and cetyltrimethylammonium bromide surfactant and tested it in a closed-loop pipeline flow system. Their experimental results showed that a stable transitional zone occurred at a higher Reynolds number range, which confirms that the creation of such complexes promoted the laminarization effect compared to

the investigated additive's drag reduction performance when tested separately.

In the present work, a solid particles—surfactant DRA complex will be formulated to test the effect of the introduction of soluble additives on the drag reduction performance of solid suspension flowing in an aqueous flow medium. Sand powder with a particle diameter of 420 μm will be used as a DRA and glycolic acid ethoxylate lauryl ether surfactant as a DRA to form complexes with the suspended solids. The drag reduction performance and stability against shear forces of the surfactant suspended solids and their complexes will be experimentally investigated and compared.

2 Methods

2.1 Materials

Glycolic acid ethoxylate lauryl ether (GCELE), also known as Laureth-4 carboxylic acid, is a non-hazardous anionic surfactant supplied by Sigma-Aldrich in liquid form. The molecular formula of $\text{CH}_3(\text{CH}_2)_{11-13}(\text{OCH}_2\text{CH}_2)_n\text{OCH}_2\text{CO}_2\text{H}$. The molecular weight of the surfactant is 739.2 kg/m³, and its density is equal to that of water (1000 kg/m³).

The sand powder (1850 kg/m³) used in this study was obtained from a local market and dried in an oven set to 60 °C for 24 h. The sand particles were separated using a sieve shaker to have the solid particles with an average particle size range of 420 μm .

2.2 Solutions preparation

The present work investigates three types of solutions: surfactant solutions with concentrations of 500, 700, 1000, and 1300 wppm, suspended solid solutions with addition concentrations of 800, 1200, and 2000 wppm, and complexes of selected samples.

Each solution is formed by blending the necessary weight of powder with water. The concentration is expressed in weight parts per million (wppm), which is calculated as follows [22]:

$$wppm = \frac{w_{\rm S}}{w_{\rm w}} \tag{1}$$

where w_s is the weight of the sand added (g) and w_w is the weight of the water (g).

2.3 Rheology test

A rheometer (Malvern Kinexus Lab+) was used to determine the rheological parameters of all solutions tested. The viscosity of the DRA solutions was evaluated at shear rates ranging from 12 to 810 s⁻¹ using the cone-plate geometry (CP2/60 SR22750SS), while the viscoelasticity characteristics were analyzed at frequencies ranging from

50 to $0.1~{\rm s}^{-1}$ using the parallel-plate geometry (PU60 SR3192 SS).

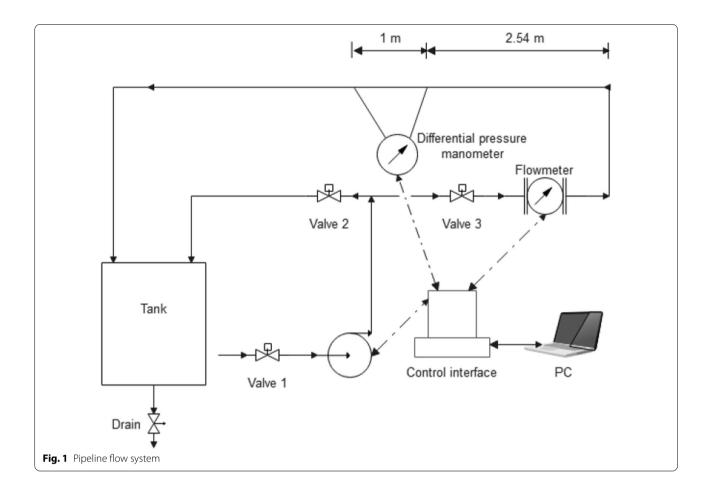
2.4 Pipeline system

A closed-loop liquid circulation system was used to investigate the drag reduction performance of the investigated solutions. Figure 1 shows the basic system design consisting of a computer, controller interface, flow meter, differential pressure gauge, centrifugal pump, valves, and a tank. The flow loop consists mainly of a storage tank (0.1 m³) connected to a straight 1-inch horizontal pipe supported with a valve to control the output. The pipe is then connected to a T-junction to create two sections. The first section is a 1-inch pipe supported with a valve that returns to the tank and is used to control the flow rate in the main testing section (second section). The testing section pipe diameter is 1 inch and is connected to a calibrated flow meter followed by an elbow that connects the entrance pipe to the main testing section. The length of the testing section was taken to be 100 times the pipe's internal diameter to ensure fully developed flow (100 inches). After the 100 inches length (2.54 m), a 1-m pressure drop measurement section is placed and connected to a calibrated differential pressure manometer (maximum error less than 0.04%). The pressure drop measurements are transferred to a SCADA system connected to a computer, and the pressure drop data collection period was 1 s.

2.5 Experimental procedure

The experiments were conducted in three phases. The first phase examined the surfactant's drag reduction performance at different addition concentrations (500, 700, 1000, and 1300 wppm). The amount for each concentration was calculated using Eq. 1 and measured and added to 1 L of water in a glass container. The solution was mixed for 2 h to ensure complete dissolution and dehydration before being introduced to the main tank. The solution will be pumped into the closed-loop liquid circulation system with different flow rates ranging from 5 to 8.4 m³/h. At each flow rate, the pressure drop measurements are taken for 20 s. This procedure is repeated at least three times for every concentration to ensure measurement accuracy, and each time a new surfactant solution is tested.

Phase two was conducted using the insoluble additive (solid powder) following the same procedure and operating parameters as in the surfactant solutions. The third phase was conducted by creating surfactant–solid complexes with the minimum and maximum concentrations by mixing the desired concentrations in a glass container for 2 h before introducing the solution to the main tank



and testing its drag reduction performance at precisely the same operating conditions.

Reynolds number (Re) was calculated using the formula in Eq. 2 [2]:

$$Re = \frac{\rho \cdot u \cdot D}{\mu} \tag{2}$$

where ρ is the solution density (kg/m³), u is the solution velocity (m/s), D is the pipe diameter (m), and μ is the solution viscosity (Pa s).

The solution velocity was calculated using the flow rate as in Eq. 3 [2]:

$$u = \frac{Q}{A} \tag{3}$$

where Q is the solution flow rate (m³/s) and A is the pipe cross-sectional area (m²).

The percentage drag reduction (%Dr) was calculated using Eq. 4 [2]:

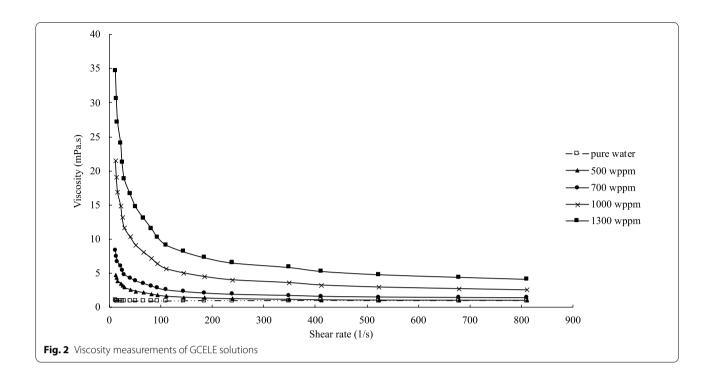
$$\%Dr = \frac{\Delta P_{\text{water}} - \Delta P_{\text{Solution}}}{\Delta P_{\text{water}}}$$
 (4)

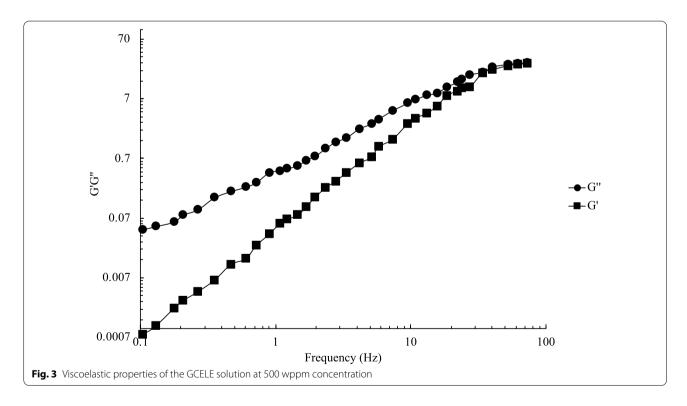
where ΔP_{water} is the pressure drop measured for pure water running at different flow rates (Pa) and ΔP_{water} is the pressure drop of the solutions running at the same flow rates as the pure water (Pa).

3 Results

Figure 2 shows the viscosity measurements of the surfactant solutions with different addition concentrations. All the investigated solutions show non-Newtonian shear thinning behavior, and the relationship between the addition concentrations and the viscosity values was linear. At low shear rates $(12-100~\rm s^{-1})$, the viscosity values increased by increasing the additive concentration with a distinguishable margin. At higher shear rates, the viscosity values declined and at low surfactant concentrations (500 and 700 wppm) reached the pure water viscosity, which indicated a breakup of the formulated micelles.

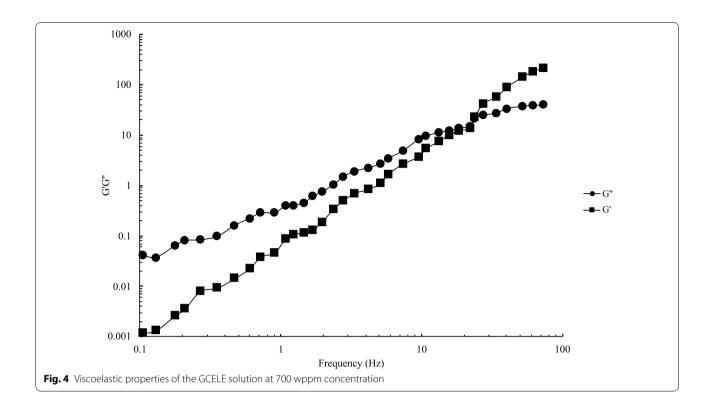
The viscoelastic properties of the surfactant solutions are shown in Figs. 3, 4, 5, and 6. All the viscoelasticity test results confirm the viscoelastic nature of the investigated surfactant solutions and agree well with the viscosity measurements presented in Fig. 2, confirming that the formulated solutions are classified as non-Newtonian

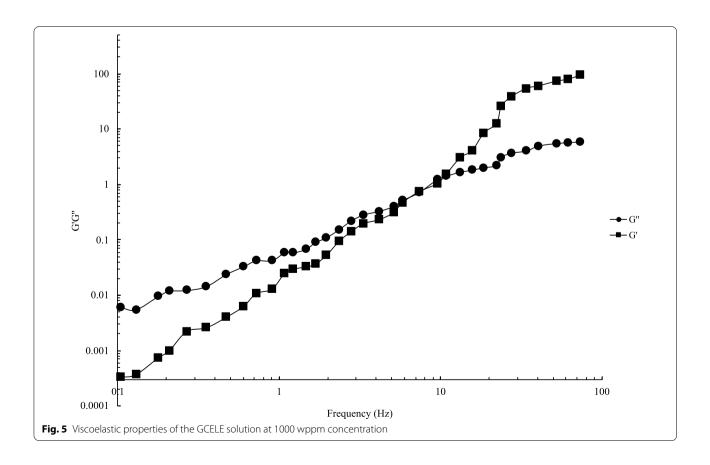


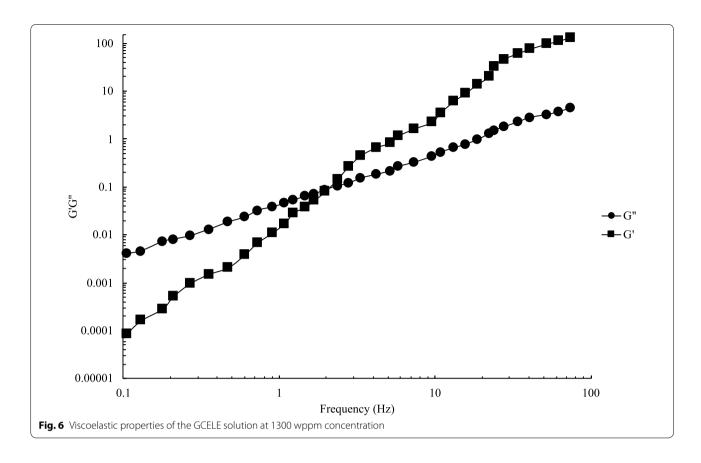


solutions. When strained, viscoelasticity tests showed the properties of a material that displays both viscous and elastic behaviors. The G' is the storage modulus which describes the elastic characteristic in which energy may

be stored in the structure during deformation. The G'' is the loss modulus which refers to the viscous behavior of the sample. When the G'' dominates the G', the solution will show a liquid-like behavior, while when the G'



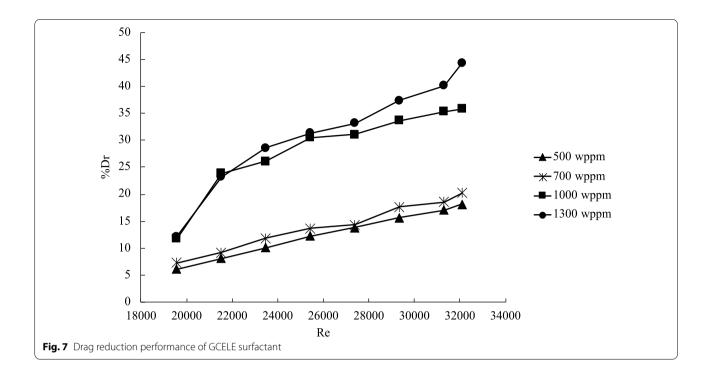




dominates the G'', the liquid will show a solid-like behavior [28]. Figures 3, 4, and 5 show the G' and G'' curves for the 500, 700, and 1000 wppm solutions, respectively. It is evident that the surfactant solutions at low concentrations (500 wppm) show a viscoelastic behavior at very high frequencies with no clear crossing between the G' and G'' curves. Increasing the concentration to 700 wppm resulted in a clearer viscoelastic behavior (Fig. 4) where the crossing between the G' and G'' curves happened when the frequency reached 34 Hz. A further increase in the surfactant concentration resulted in a firmer viscoelastic behavior (Fig. 5) with earlier crossing at the frequency of almost 10 Hz. Figure 6 shows the viscoelastic properties of the 1300 wppm surfactant solution where the cross between the G' and G'' line occurred at a very low frequency (2.2 Hz).

Figure 7 shows the effects of the degree of turbulence and the additive concentration on the %Dr of the GCELE surfactant. The results show that the GCELE surfactant is an effective DRA with a maximum %Dr of 44.3% at Re of 32,083.8. The %Dr was found to increase by increasing the degree of turbulence which is expected since the relationship between the degree of turbulence, and the additive's drag reduction performance is linear at a certain degree of turbulence (Re) ranges. Increasing the Re

means increasing the number of turbulence structures in the main flow, and that will provide a suitable flow environment for more effective interaction between the DRA and the whole flow, resulting in a higher degree of turbulence suppression. It is known that the ideal behavior of soluble additives will consist of four main responses depending on the Re value and the operating conditions. The three zones will be the very low %Dr zone where the degree of turbulence is not enough to enable the additives to suppress turbulence, the onset point zone where the degree of turbulence is enough to start effective interaction with the additives, and the drag reduction zone where the additive reaches its maximum drag reduction performance and the declining zone where the additives lose all or part of their drag reduction efficiency [20, 27]. The fourth zone was not observed in the present work where no decline in the %Dr was spotted, and that means the investigated Reynolds number range in the present work was not enough to overcome the surfactant drag reduction effect. The figure also shows the effect of the surfactant concentration on the drag reduction performance. The figure shows clearly that the %Dr increases by increasing the surfactant concentration, which is expected since increasing the surfactant concentration means increasing the number of molecules contributing

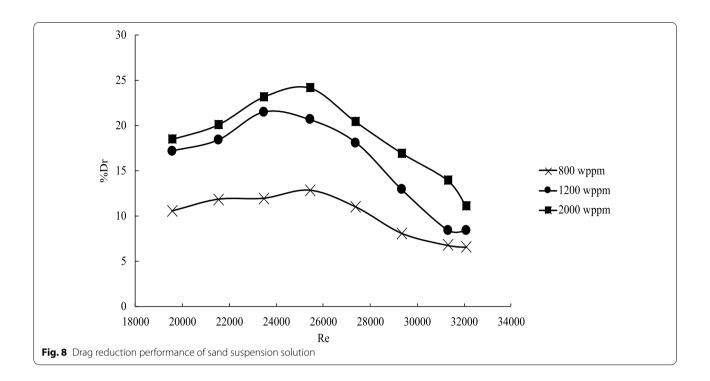


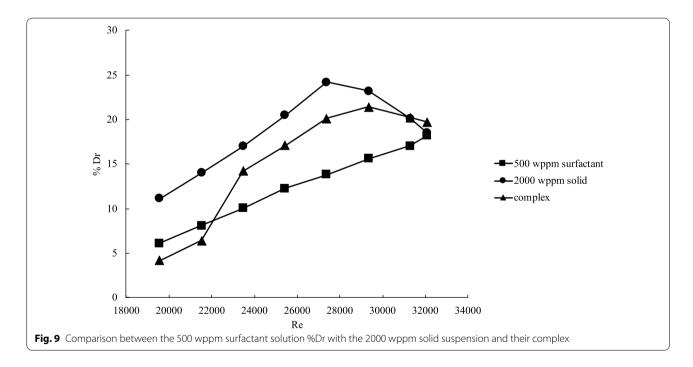
to the whole drag reduction system. Interestingly, the increase in the %Dr was not linear where the %Dr values of the 500 and 700 wppm were close, and the differences between them and the 1000 and 1300 wppm were high. Such behavior clearly indicates the complex drag reduction phenomena and its relationship to all the controlling parameters that act simultaneously and in the same flow medium. It is believed that at low additives concentrations, the number of surfactant molecules was enough to act as turbulence suppressors on the smaller eddies forming the turbulent flow, and the medium size and large eddies were not affected. Increasing the additive's concentration enabled the presence of more surfactant molecules to suppress small- and medium-size eddies with part of large eddies, which are believed to dominate the flow system.

The drag reduction performance of the sand suspension solutions is shown in Fig. 8. The results indicated that the sand particles within the particle size range investigated can act as an effective DRA. The figure shows that the relationship between the %Dr and Re is much more pronounced than the surfactant solutions result, where the inclined–maximum–declined behavior is more evident. A maximum %Dr of 24.4% was achieved at Re=25,431 and 2000 wppm concentration. Contrary to the surfactant solutions, the sand suspension behavior was almost identical to the traditional behavior of many known additives, and it is believed that the type and addition concentrations contributed significantly to

the turbulence-additives interaction relationship discussed earlier in the GCELE drag reduction responses to the increase in the degree of turbulence. The mechanism controlling the drag reduction performance using insoluble additives is different. It mainly focuses on the solid particle's interaction with the eddies and the effect of the solid properties such as the particles diameter, density, and particle shape on the overall shape drag reduction behavior. It is clear from the figure that the degree of turbulence at low Re ranges was enough to create an effective suspension that can suppress the eddies and enhance the flow reaching the maximum %Dr at Re=25,431. A further increase in the Re (degree of turbulence) resulted in a dramatic loss in the drag reduction performance, reaching levels even lower than that at low Re. It is believed that at high Re ranges, the degree of turbulence will be higher and that will overcome the eddies suppression effect of the DRA. In other words, when the additive interacts with the formed eddies, new viscoelastic properties will be introduced and that will result in suppressing the eddies and preventing them from completing their shape. An increase in the degree of turbulence (Re) will create an intense shearing flow environment that overcomes the viscoelastic additive's effect which will result in a reduction in the additive's drag reduction efficiency.

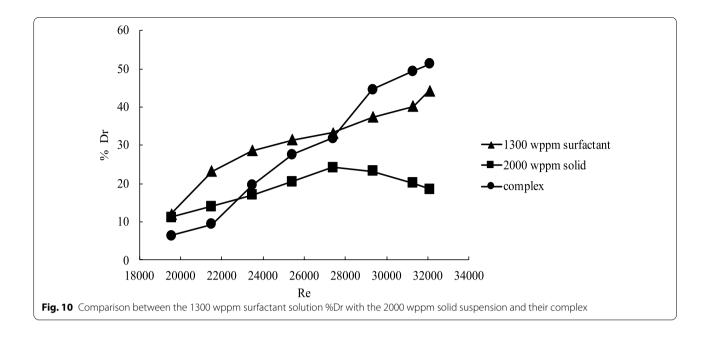
The drag reduction performance of the surfactant-solid complexes and their comparison with their initial solutions are shown in Figs. 9 and 10. The





surfactant–solid complexes are formed by mixing the exact amounts of surfactant and solid used to create their solutions. Figure 9 shows that the surfactant–solid complex (500 wppm surfactants + 2000 wppm solid) drag reduction performance was lower than the surfactant and the solid at low Re (19,563–21,519).

Such low values of the %Dr indicated that the complex is successfully formed, and its viscoelastic properties were different from that of the surfactant, which resulted in higher resistance to low shearing rates at a low degree of turbulence. Increasing the Re resulted in increasing the %Dr to be higher than the solid and



lower than the surfactant reaching up to the maximum Re, where a slight decline in the %Dr of the complex was observed and almost identical %Dr for the three investigated solutions. It is believed that the presence of the suspended solids in the surfactant network had an enforcing effect that increased the suspended solid resistance to high shear forces applied by the turbulent structures (eddies). Also, the presence of the suspended solids enhanced the surfactant drag reduction efficiency by 40–44% at Re ranging between 21,519 and 31,300, which indicates the creation of surfactant-enforced aggregates with high turbulence suppression efficiency.

Almost the same behavior was observed with surfactant-solid complexes at maximum concentrations (1330 wppm surfactant + 2000 wppm solid), as shown in Fig. 10. The difference, in this case, is that the complex drag reduction performance was higher than the two individual additives, and the maximum %Dr was not reached. In this case, increasing the surfactant and solid concentrations in the formed complex created an enforced complex with high resistance to shear forces and better interaction and eddies suppression effects that dominated the other two individual solutions' performances. A maximum %Dr of 52% was observed with the 1300-2000 wppm complex which is higher than the maximum %Dr observed for the two individual solutions which indicates a clear enhancement in the drag reduction performances by the creation of the complexes.

4 Discussions

Surfactants, in general, can act as drag reducing agents in pipelines transporting commercial liquids in turbulent flow mode due to their polar nature which enables them to form certain kinds of aggregated (micelles) that can act as one unit. The polymeric additive's resistance to high shear forces is low and can result in an irreversible degradation that will noticeably reduce their drag reduction performance. On the contrary, after exposure to high shearing forces, surfactant polar nature will enable the reformation of the micelles and regaining their drag reduction abilities. Despite that, the surfactants are not implemented commercially due to their low drag reduction performance when compared to high molecular weight polymers. In the present work, enforcing the surfactant micelles with solid particles was experimentally proven to enhance the drag reduction performance of the surfactant solutions within the applied experimental conditions. The surface of the sand particles within the investigated particle size (420 µm) is positively charged since the particle's diameter is more than 250 µm as indicated by Gu and co-workers [8]. The oppositely charged surfactant molecules will be attracted and attached to the surface of the suspended sand particles forming solidenforced micro-aggregates that have the viscoelastic nature of the surfactant molecules and the rigidity of the solid particles.

The traditional interaction between the turbulence structures (eddies) and the additives is the key factor in reducing the drag by suppressing the eddies themselves.

This happens when new viscoelastic properties are introduced to the structure of the eddy which prevents it from forming the same shape and the same size as in the additives-free flow systems. The surfactant-solid complex investigated has higher resistance to the eddy shearing forces and that resulted in higher drag reduction performances as presented in Figs. 9 and 10. It is interesting to see that the drag reduction performance of the surfactant-solid complex is not always higher than the individual solutions (surfactant and sand suspension) within the investigated Reynolds number (Re) range. To explain this point, it is important to highlight the relationship between the degree of turbulence, represented by Re, and additives efficiency. The relation between the degree of turbulence and the drag reduction efficiency of the additives has three major modes which are the onset point (the first turbulence suppression point), the maximum drag reduction point, and the decline mode (when the degree of turbulence overcomes the efficiency of the additives). Figure 10 shows that the three modes are observed when testing the sand suspension, while the decline mode was not observed for the surfactant solution and the complex. Also, tracing the complex curve trajectory shows that it had the lowest %Dr at Re range of 19,563-23,475, while it was higher than the solid at Re range of 23,475-27,388 and finally showed a higher %Dr than the surfactant solution at Re range of 27,388-32,083. Such important observation suggests that the formed surfactant-solid complex elevated the optimum interaction point (optimum drag reduction point) to be at a higher degree of turbulence which indicates the formation of a hybrid surfactant-solid aggregate with higher drag reduction performance and high resistance to mechanical degradation.

5 Conclusions

The present work aims to investigate the drag reduction performance of a new surfactant-solid complex created from glycolic acid ethoxylate lauryl ether (GCELE) and sand particles with a particle size of 400 µm. The drag reduction performance of the surfactant, solid, and surfactant-solid complexes solutions was tested in a closed-loop liquid circulation system with different addition concentrations and different flow rates. Also, the rheological and viscoelastic properties of surfactant solutions were investigated to indicate the solution's non-Newtonian nature. The experimental results showed that all the surfactant solutions exhibited shear thinning non-Newtonian behavior. The surfactant's drag reduction performance was higher than the suspended solids that exhibited traditional incline and declined their %Dr values due to the absence of the viscoelastic characteristics as in the surfactant solutions. The formulated complex's drag reduction performance was higher than the surfactant one when created in a low concentration mixture (500 wppm surfactants + 2000 wppm solid), indicating the creation of surfactant-solid-enforced aggregates in the main flow that have higher resistance to the turbulence shear forced and better turbulence suppression efficiency. On the other hand, the surfactant-solid complex created with the maximum concentrations (1330 wppm surfactant + 2000 wppm solid) showed a higher %Dr compared to the individual solutions of the surfactant and solid and without reaching the maximum %Dr point. Such behavior is related to the formation of surfactantsolid aggregates that act as one large rigid molecular network with high viscoelastic properties and density, and this network can suppress turbulence eddies at a high degree of turbulence.

Abbreviations

 $\Delta P_{solution}$: Pressure drop of solutions (mbar); ΔP_{water} : Pressure drop of pure water (mbar); %Dr: Percentage drag reduction (%); A: Pipe cross-sectional area (m²); D: Pipe internal diameter (m); Q: Solution volumetric flow rate (m³/h); Re: Reynolds number; u: Solution velocity (m/s); w: Solution concentration (weight parts per million); w; Weight of the sand added (g); w,: Weight of the water (g); μ : Solution viscosity (Pa.s); ρ : Solution density (kg/m³).

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Author contributions

WK carried out the experimental work, solutions preparation, and contributed to the data analysis and manuscript writing. MH contributed to data analysis and manuscript writing. HA designed and build the closed-loop flow system and contributed to the data analysis and manuscript writing. All authors read and approved the final manuscript.

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