

A new low interfacial tension viscoelastic surfactant for EOR applications in low permeability reservoirs

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Received: May. 06, 2020; Accepted: July. 27, 2020

The development of low permeability reservoirs has become the main battlefield for oil field exploitation. However, due to its special and complicated geological conditions, the existing chemical enhanced oil recovery methods cannot meet the needs of the efficient development of low permeability reservoirs. Herein, the low interfacial tension viscoelastic surfactant (GACS) was studied as the potential candidate for low permeability reservoir oil displacing agent. The parameters characterizing the efficiency of chemical flooding for enhanced oil recovery were studied: surface activity, viscosity, oil-water interfacial tension (IFT), and viscoelasticity. The results show that, as a new class of chemicals for EOR under harsh conditions of high salinity, the GACS is made up of a single component, are shear-thinning with good injectivity. The critical micelle concentration (CMC) and surface tension of GACS surfactants are extremely low, which is conducive to the formation of micelles at low concentrations. Under different salinity conditions, the IFT is at the low to ultra-low interfacial tension level, and the surfactant solution demonstrated remarkable viscosifying ability and viscoelasticity. GACS was found to be a potential additive for the enhanced oil recovery application.

Keywords: EOR applications; CMC; low interfacial tension; viscous property; viscoelastic property; Gemini surfactant.

[http://dx.doi.org/10.6180/jase.202012_23\(4\).0015](http://dx.doi.org/10.6180/jase.202012_23(4).0015)

1. Introduction

Low permeability reservoirs account for significant oil sources worldwide. According to reports [1–3], approximately 38 % of oil and gas in the world is largely recovered from low permeability reservoirs. Almost 46 % of produced oils in China are recovered from low permeability reservoirs. Concomitantly, recovered oil from low permeability reservoirs amounts to more than 50 % of produced oils in the US. Owing to the unceasing development of the world economy, increasing oil demands are inevitable. Thus, rational studies for recovering oil and gas from low permeability reservoirs present imperative perspectives in the petroleum industry. The exploration, development, and feasible recovery of oil from low permeability reservoirs have been often a difficulty and frontier subject for production engineering in the industry [3, 4].

At present, low-permeability reservoirs usually use water flooding to improve oil recovery, but the technology of enhanced oil recovery after water injection is still in its infancy. Chemical enhanced oil recovery (CEOR) is generally applied in mature assets that have been water flooded over a long period and have a significant water cut and it is an exceptional and economically feasible method for improving oil recovery [2, 3, 5]. However, low permeability reservoirs are endowed with tight reservoirs, complex pore structures, strong heterogeneities, and serious Jamin effects [3, 6]. Due to low permeability reservoirs' special and complicated geological conditions, many conventional CEOR techniques such as polymer flooding, surfactant flooding, surfactant-polymer (SP) flooding, and alkali-surfactant-polymer (ASP) flooding are unsuccessfully applied [7, 8]. In summary, the production of oil from low permeability reservoirs is associated with several difficulties, and the existing EOR

techniques cannot meet the requirements. Accordingly, it is necessary to seek for new technical scheme to improve oil displacements in low permeability reservoirs, hence enhancing oil recovery.

In general, there have been two major approaches used in CEOR: 1) increase the viscosity of injected water with the water-soluble polymer to increase the mobility ratio between the displacement fluid and crude oil, thereby increasing the volume sweep efficiency or 2) adding surfactants to reduce the interfacial tension (IFT) between oil and water, allowing the trapped oil to flow through tight pore necks [5, 9].

Viscoelastic surfactants are a class of surfactants that form a micelle structure in aqueous solution and have viscoelastic properties, and have broad application prospects in fracturing, acidification, drilling fluid, improving recovery rate and fluid reduction and transport [10–18]. When the concentration of the viscoelastic surfactant solution increases to a certain critical concentration, the spherical micelles formed by self-assembly in the solution begin to transform into worm-like micelles, and the viscosity of the solution increases. As the concentration further increases, worm-like micelles grow rapidly and form flexible rod-like micelles. At the same time, flexible rod-shaped micelles are entangled and adhered to each other under the action of applied shear force and molecular thermal motion to form the supramolecular three-dimensional network structure. The viscosity and elasticity of the solution are becoming more and more obvious, and the rheological properties are close to Newton viscoelastic fluid. Worm-like micelles have always been in a dynamic balance of winding and dispersion, reorganization and fracture, so the fluid viscosity does not change with time at a certain shear rate [19–23]. The reversible formation and deformation of micelles aids easier and non-damaging propagation in heterogeneity reservoirs, hence bettering oil swept efficiency. Moreover, viscoelastic surfactants superbly reduce the oil-water interfacial tension and may alter the reservoir wettability desirably, causing substantial betterments for oil displacement efficiency [8]. Therefore, viscoelastic surfactants represent the most promising amphiphilic compounds that possess a combination of desirable properties for the EOR process i.e. IFT reduction and fluid viscosifying properties [24, 25], and it can effectively improve the recovery of low permeability reservoirs, and some scholars have conducted related research.

In 2007, Istvan Lakatos et al. [26] first evaluated viscoelastic surfactants as mobility controlling agents used as pre-flush, co-surfactant, and post-buffer media. Studies have shown that viscoelastic surfactants can replace tradi-

tional mobility control agents (polymers) in a wide range of temperatures and pressures. Therefore, viscoelastic surfactants can provide excellent opportunities for fluidity control in various enhanced oil recovery technologies independent of injection schemes, and can effectively increase recovery rates by 10 %. These studies have opened up new prospects for the application of viscoelastic surfactants in chemical flooding to improve recovery. L. Siggel et al. [5, 9] reported a new class of viscoelastic surfactants and their EOR potential in high temperature and high salinity reservoirs. And developed TPM series viscoelastic surfactants, of which TPM-101-10 viscoelastic surfactant has the best performance. It has viscoelasticity at low concentration (<0.5 % w/w) and shows good stability in mineralized water containing a high concentration of divalent cations at a high temperature (> 70°C) and high salt (186 g/L TDS). It has acceptable adsorption values on sand and clay and shows good injection capacity in Darcy Gildehaus sandstone. In the follow-up study, the researchers conducted core flooding experiments on TPM-101-10. TPM-101-10 viscoelastic surfactant mobilize residual oil (ca.7 % OOIP). Zhu et al. [8] proposed the use of small molecular viscoelastic surfactants in porous media with special rheology and good interfacial activity to improve oil recovery in low permeability reservoirs. In response to the reservoir conditions, the researchers chose a zwitterionic betaine surfactant with a long carbon chain, EAB, which exhibits viscosity-increasing behavior, shear-thinning characteristics, low IFT performance (10^{-3} - 10^{-2} mN / m) and good injectability. The incremental oil recovery of single viscoelastic surfactant formula flooding (0.4wt % EAB) is 12.6 %. The viscoelastic surfactant flooding may have great potential in EOR applications in low permeability reservoirs. Fan et al. [27] prepared the new type of viscoelastic surfactant solution with high salinity tolerance and fast-dissolving performance to enhance oil recovery in offshore oilfields. In this work, a new type of viscoelastic surfactant solution, which meets these criteria, was prepared by simply mixing the zwitterionic surfactant HDPS or ODPS with anionic surfactants such as SDS. This is a combination of surfactants and not a single viscoelastic surfactant.

In order to facilitate easier and economically feasible EOR operations in low permeability reservoirs, we have carried out relevant research works for viscoelastic surfactants [28–31]. In those studies, we developed and investigated a potential viscoelastic surfactant for the oil and gas industry. The viscoelastic surfactant displayed outstanding rheological properties which are the most desirable requirements for EOR through surfactant flooding. Despite the exceptional performances, the studies were merely limited

for fracturing drilling perspectives. Besides, the studies were limited to pure water giving no reflections in the actual oilfields which are naturally saline vicinities. In the present work, we investigate the potentiality of the developed viscoelastic surfactant for EOR operations. To reflect the actual performance in the oilfields, simulated oilfield conditions were applied. The simulated field condition is a condition of an oil field in China, and the selected block is a low-permeability reservoir.

2. Experimental

2.1. Materials

The viscoelastic surfactant (coded: GACS), Fig. 1, was obtained from the laboratory of the School of Petroleum Engineering, Yangtze University. Crude oil and formation water for an oil field in China and the properties are shown in Tables 1 and 2, respectively.

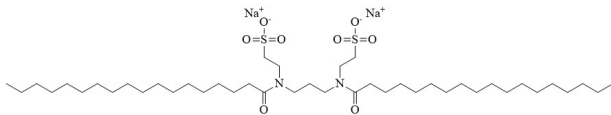


Fig. 1. Chemical structure of GACS.

2.2. Solution preparation

The formation of water was filtered using filter paper (45 μm). Then water with varying salinities was prepared by mixing distilled water and formation water different proportions, Table 3. The GACS solutions with different concentrations were prepared using synthetic water. The aqueous solutions of GACS were sonicated for sufficient time to ensure complete mixing.

2.3. Measurements

2.3.1. Surface tension

The surface tension of solution at the air-aqueous surfactant interface was recorded by a QBZY type automated surface tensiometer at room temperature (27°C). Each surface tension measurement was repeated three times. The CMC values were determined by the graphical method as we reported in our previous work [29].

2.3.2. Viscosity

The viscosity measurements were studied by DV-IIT Brookfield viscometer. The viscosities of the solutions (0.1wt.% to 0.5wt.%) at a shear rate of 7s^{-1} were recorded at a temperature range of 30°C to 60°C. The viscosity data were all averaged three times, and the viscosity test error was about ± 0.005 mPa·s.

2.3.3. IFT

The oil-water interfacial tensions between crude oil and surfactant solutions were measured by using Texas-500 spin drop interfacial tensiometer. All the measurements were conducted at 45°C which is the expected reservoir temperature for the oilfield. The spinning oil droplet in the surfactant solution was stretched until the oil-water phase attained equilibrium at a rotation speed of 5000 r/min.

2.3.4. Viscoelastic

Viscoelastic property of the surfactant solution was studied by measuring the modulus (i.e. G' , and G'') using an MCR 301 Rheometer at 45°C. The linear viscoelastic region of the solution was first determined by stress scanning. The strain scanning range was 0.01 % to 100 % and the frequency was 1Hz. The change of modulus with shear strain was investigated to determine the linear viscoelastic region of the solution. In the linear viscoelastic region, the solution was frequency scanned, and the angular frequency was changed from 0.1 to 100 rad/s to determine the relationship between the storage modulus G' and the loss modulus G'' and the oscillation frequency. According to the change of G' and G'' of the viscoelastic fluid with the oscillation angular frequency, the loss factor $\tan \delta$ and the relaxation time τ were calculated to evaluate the viscoelasticity of the solution. Calculation formulas as follows:

$$\tan \delta = \frac{G''}{G'} \quad (1)$$

$$\tau = \frac{2\pi}{\omega} \quad (2)$$

3. Results and discussion

3.1. Surface activeness

The surface tension of the GACS surfactant solution at different salinities was investigated by changing the concentration, see Fig. 2 and Table 4. As is shown in Fig. 2, the surface tension of the solution gradually decreases as the concentration increases. After reaching a certain value, the surface tension almost no longer changes. The critical micelle concentrations (CMC) of the surfactants can be determined from the inflection point in the curve. Surface activity test shows that under different conditions of salinity, the CMC of GACS surfactant is in the range of 0.02 ~ 0.05 mmol/L, which means that at very low concentration, the solution can self-assemble to form the micelle structure, which it is beneficial to increase the viscosity of the solution and exert its excellent viscoelasticity during the oil displacement process. As the salinity increases, surface activity increases and CMC decreases. This is due to the elec-

Table 1. Properties of crude oil and reservoir temperature for oilfield.

Temperature (°C)	Density (g/cm ³)	Viscosity (mPa.s)	Formation oil volume factor	Gas-oil ratio (8.93m ³ /t)
45	0.82	4.62	1.04	8.93

Table 2. Water chemistry of formation water for oilfield.

pH	Ion concentration (ppm)							Salinity (ppm)	Water type
	Na ⁺ +K ⁺	Ca ⁺⁺	Mg ⁺⁺	Cl ⁻	SO ₄ ²⁻	CO ₃ ²⁻	HCO ₃ ⁻		
6.5	8633	1083	129	12064	2546	38	383	24855	CaCl ₂

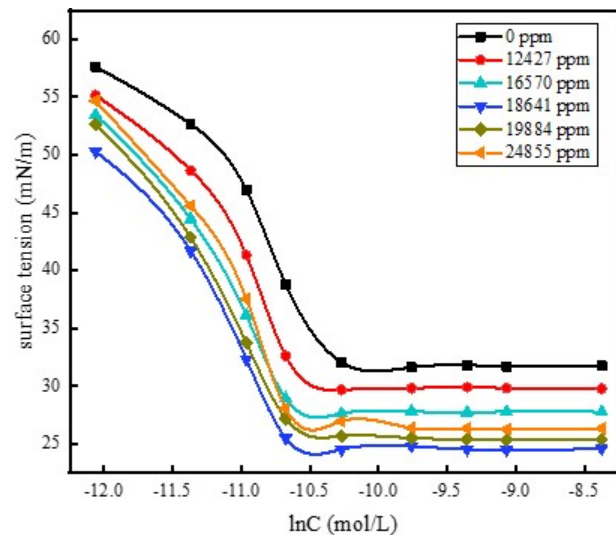
Table 3. The salinities for synthetic waters used in the study.

Synthetic water	Distilled water	Synthetic water1	Synthetic water2	Synthetic water3	Synthetic water4	Formation water
Water mixture (distilled water:formation water)	1:0	1:1	1:2	1:3	1:4	0:1
Salinity (ppm)	0	12427	16570	18641	19884	24855

trostatic interaction between the positively charged cations and the negatively charged anion groups after the addition of inorganic salts to the aqueous solution of Gemini surfactants. Due to the presence of electric charge, there is an electric double layer at the interface between micelles and water, and the water molecules form the hydration membrane. The addition of inorganic salts destroys the hydration membrane around the micelles, and also compresses the diffusive electric double layer around the ionic groups, shielding the repulsive force between the charges, making the surfactant molecules arranged closer together, and surface active rise [32–35]. However, under high salinity conditions, surface activity decreases and CMC increases. This is because the inorganic salt shields the repulsive force between the polar heads and also shields the electrostatic attraction. When the salinity is small, the repulsion between the ionic groups dominates. But with the increase of salinity, the force of electrostatic attraction between ions increases, and salt shields the electrostatic attraction between ionic groups, thereby weakening the interaction between the two and reducing the surface activity [36–39].

3.2. Viscosity

The viscosifying ability of the surfactant is another imperative property to be considered during the selection of surfactants for EOR operation [40]. The viscous property principally controls the mobility of the fluid system and hence the macroscopic swept efficiency. The effect of concentration on solution viscosity under different salinity conditions at reservoir temperature (45°C) was investigated, see Fig. 3. The surfactant concentration affects the micellization of surfactant molecules in the bulk phase, thereby

**Fig. 2.** Surface tension of GACS surfactant at different salinities.

affecting the viscosity properties of the solution [41]. Under the same salinity condition, as the concentration increases, the viscosity of the solution increases. This phenomenon can be explained by the tendency to form different layered micelles with the change of surfactant concentration. The surfactant forms dense entangled networking micelles with increasing concentration which is beneficial to the viscosifying effect [28]. According to the test results, when the concentration of the solution is 0.5wt.%, the viscosity of the solution can be greater than the viscosity of the formation crude oil under various salinity conditions (two-dot chain line in the figure, 4.62 mPa.s). The effect of temperature

Table 4. Surface activity properties of GACS surfactant at different salinities.

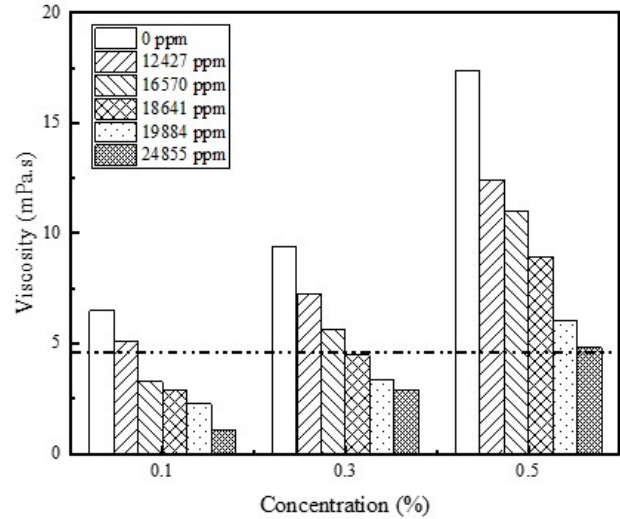
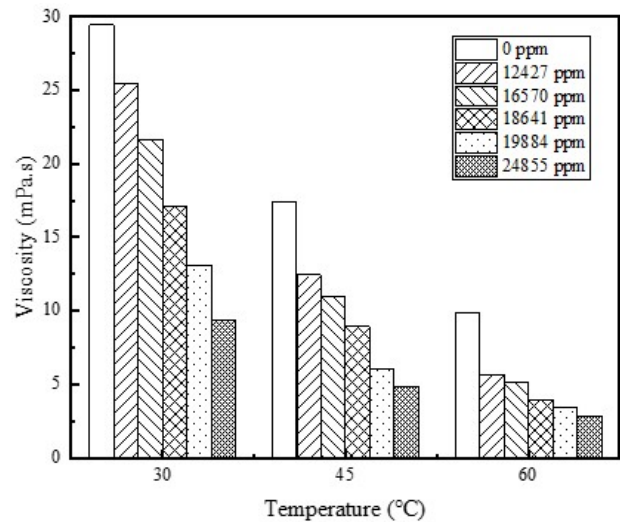
	Salinity (ppm)					
	0	12427	16570	18641	19884	24855
CMC (mmol/L)	0.0407	0.0328	0.0271	0.0262	0.0268	0.0283
γ_{CMC} (mN/m)	31.6826	29.7922	27.7023	24.6434	25.662	26.8052

on the viscosity of 0.5wt.% GACS solution under different salinity conditions was investigated, as shown in Fig. 4. When the salinity of the solution is higher than 20000 ppm, a severe decrease in viscosity is observed. Temperature elevation also diminishes the viscosity of solution under the same salinity. All those observations are explained from the micelle formation viewpoint. GACS is well-known to self-assembly in solution forming entangled layer-like micelles which are responsible for viscosifying the solution [30, 42]. The increase in salinity presumably reduces the interaction between the micelles themselves and other components. The reduced interaction weakens the entanglement of micelles which provoked the movement of micelles within the solution and consequently diminished viscosity. When the salinity is high enough (salinity greater than 20000ppm), GACS molecules ostensibly precipitate resulting in disintegrated micelles. The disintegration of micelles illustrates the phenomenon that the viscosity of the solution drops significantly at higher salinity values. In general, temperature elevation triggered the movement of micelles through weakening the entanglements of micelles; thereby reducing the viscosity of the solution when the temperature increases. Despite the adverse effects of temperature, the surfactant solution still exhibits significant viscosity at 60°C. The formation of micelles ensures viscosity at moderate temperatures.

Despite those decrements in viscosity of solution due to salinity increase and temperature elevation, 0.5wt.% GACS substantially viscosified the solution even at high salinity and relatively high temperatures. The viscosity of GACS surfactant is much higher than the reported TPM (1 ~ 10 mPa.s) [9], VES-JS system (0 ~ 16 mPa.s) [43], VES system (3 mPa.s) [44, 45], equivalent to HDPS/SDS system (42.3 mPa.s) and ODPS/SDS system (23.8 mPa.s) [27]. In general, GACS has a remarkable viscous property and may practically substitute the polymeric viscosifiers which are often reported to cause secondary formation damage with an imperative property of more interfacial activity [4, 28].

3.3. Interfacial tension

To facilitate enhanced oil recovery, the oil surfactant - water interfacial properties of an interface is the most relevant [46]. One of the most common and simple measurement parameters of interface behavior is the interfacial tension

**Fig. 3.** The effect of concentration on the viscosity of GACS solution at different salinities.**Fig. 4.** The effect of temperature on the viscosity of GACS solution at different salinities.

between crude oil and surfactant. For the displacement of crude oil in the pores and capillaries of petroleum reservoir rock, it is generally required to reduce the IFT to the low or ultra-low level (10^{-2} mN/m ~ 10^{-3} mN/m).

The experimental study of the dynamic interfacial tension behavior between crude oil and surfactant GACS is

shown in Fig. 5. This research not only provides information on the adsorption rate of surfactant molecules, but also provides information on the adsorption mechanism of surfactant molecules, which helps to reveal the factors that affect the adsorption process [46]. The results showed clearly that the IFT decrease with time and eventually equilibrate. The adsorption of the GACS molecules at the oil-water interface initially increases with time, causing the reduction in IFT before saturating the interface. To achieve the equilibrium between the oil and water phases, the adsorbed GACS molecules are desorbed into the bulk phase. Therefore, the saturation of GACS molecules at the interface decrements causing an increase in IFT and remains fairly the same after equilibration [29, 43]. As shown in Fig. 5, the final oil-water IFT first decreases and then increases with the increase of salinity, but both are lower than the IFT value of distilled water. Mixing pure water with formation water at the salinity of 12427 ppm is found to be the best water formulation for maintaining ultra-low IFT. At this salinity, the minimum and equilibrium IFT values are 3.49×10^{-4} mN/m and 2.93×10^{-3} mN/m, respectively. Besides, the GACS is capable to maintain ultra-low IFT (10^{-3} mN/m) up to salinity level of 18641 ppm beyond which it maintained low IFT (10^{-2} mN/m). The excellent interface activity of GACS surfactant is far superior to the reported TPM (1.2 mN/m) [5], TDPS (0.3 ~ 0.5 mN/m) [20], equivalent to ODPS/SDS system (0.01 ~ 0.1 mN/m) [27], VES-JS system (0.01 ~ 0.1 mN/m) [47], VES system (0.04 ~ 0.07 mN/m) [44, 45].

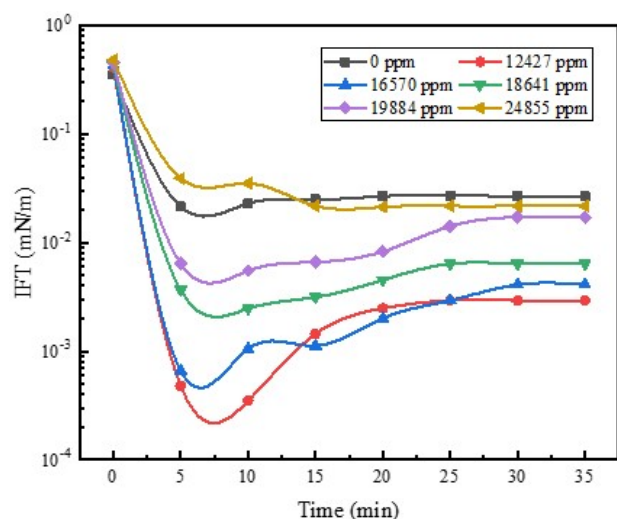


Fig. 5. Dynamic oil-water interfacial tension of GACS solution at different salinities.

The trend of IFT decreasing first and then increasing is mainly due to: after adding appropriate amount of salt, the

ionic strength of the surfactant system increases, the degree of hydration of the polar group weakens, and the diffusing electric double layer near the hydrophilic head group is compressed. This not only shields the repulsion between ionic groups, but also destroys the hydration membrane. Oil molecules and water molecules are easy to infiltrate into the surfactant, the space between the oil phase and water phase increases, the thickness of the interface layer increases, the surfactant becomes more oil-wet, which promotes their coalescence, thereby increasing the potential for distribution into the oil [35, 38, 48]. Therefore, under a certain degree of salinity, surfactants can effectively aggregate at the oil-water interface and achieve the adsorption-desorption balance. The arrangement of the Gemini surfactant adsorbed on the interface is more conducive to reducing the interfacial tension, thus reducing IFT to the level of low or ultra-low interfacial tension. However, as the concentration of salt ions increases, the repulsive force and electrostatic attraction between the polar head groups are shielded, the degree of bending and agglomeration of the surfactant film gradually increases, and the thickness of the oil-water interface layer decreases. The competition for the adsorption of surfactants and inorganic ions at the oil-water interface is strengthened, resulting in a decrease in the amount of surfactant adsorption at the oil-water interface. Therefore, the interaction between the surfactant and the oil phase gradually decreases, the solution interface activity decreases, and the IFT increases [35, 49].

3.4. Viscoelasticity

The viscoelastic nature is said to aid microscopic sweep efficiency due to the distinctive flow characteristic of the solution. The better the viscoelasticity is, the more residual oil is carried and consequently better oil displacement efficiency [43, 50]. Following the results discussed for viscosity and IFT analysis, the viscoelastic property of GACS was investigated only for the solution salinity of 0 ppm, 12428 ppm, 18641 ppm, and 24855 ppm. Experimental results for the analyses are depicted in Fig.6 (6a-6d) and 7 (7a-7d). Fig.6 (6a-6d) depicts moduli i.e. G' and G'' as the functions of shear strain. When the strain is low, G' is always greater than G'' , and the value of the two does not change significantly, showing a flat straight line. When the shear strain exceeds a certain value, G' decreases rapidly and is less than G'' , eventually exceeding the linear viscoelastic zone. Meanwhile, Fig.7 (7a-7d) shows the variation of modulus with angular frequencies. The modulus both increase with an increase in oscillation frequencies and they exhibit a crossing point/frequency. At lower frequencies, G'' is greater than G' . At higher frequencies, G' is greater

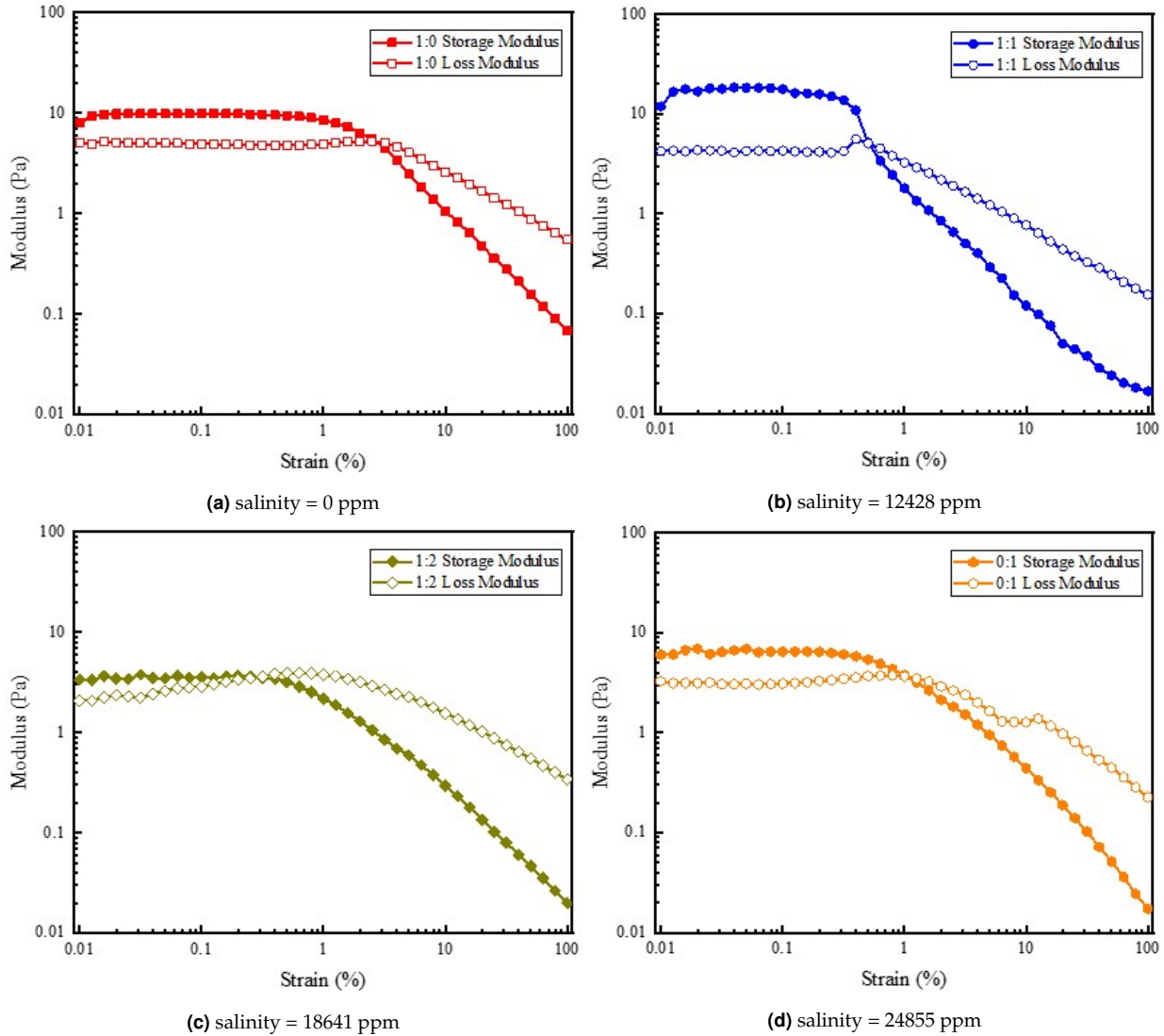


Fig. 6. Relationship between modulus and strain of GACS solution.

than G'' . This phenomenon implies that GACS solutions are more viscous at low frequencies and are more elastic at high frequencies. Conclusively, the ability of GACS solutions to switch their property from viscous to elastic after a specific cross strain/frequency confirms the viscoelastic nature of GACS solutions. The viscoelastic nature of solutions is attributed to the ability of GACS to self-assemble in solutions forming micelles which can be disintegrated and regenerated under varying stresses [43].

The $\tan \delta$ and τ results are presented in Table 5 and show that the GACS solution is mainly elastic ($\tan \delta < 1$) and exhibit a considerable relaxation time. With the increase of salinity, $\tan \delta$ of GACS solutions gradually decreases first and then increases (0.5375, 0.3661, 0.5361, 0.6528), and τ gradually increases first and then decreases (3.0784s,

7.1382s, 1.7749s, 1.1320s). This indicates that the viscoelasticity of the solution increases first and then decreases as the salinity of the solution increase. At the salinity of 12428 ppm, the solution shows the optimal elastic property and relaxation time. Currently related to EOR viscoelastic surfactants, such as VES-JS system [43] and NaOA/ Na_3PO_4 system [51], the relaxation time τ of the system is only about 1s. In summary, the values of calculated $\tan \delta$ and τ both $\tan \delta$ and τ both emphasize the promisingly viscoelastic nature of GACS which is beneficial in EOR operation through surfactant flooding.

4. Conclusions

The low interfacial tension viscoelastic surfactant (GACS) was successfully investigated for its suitability in EOR op-

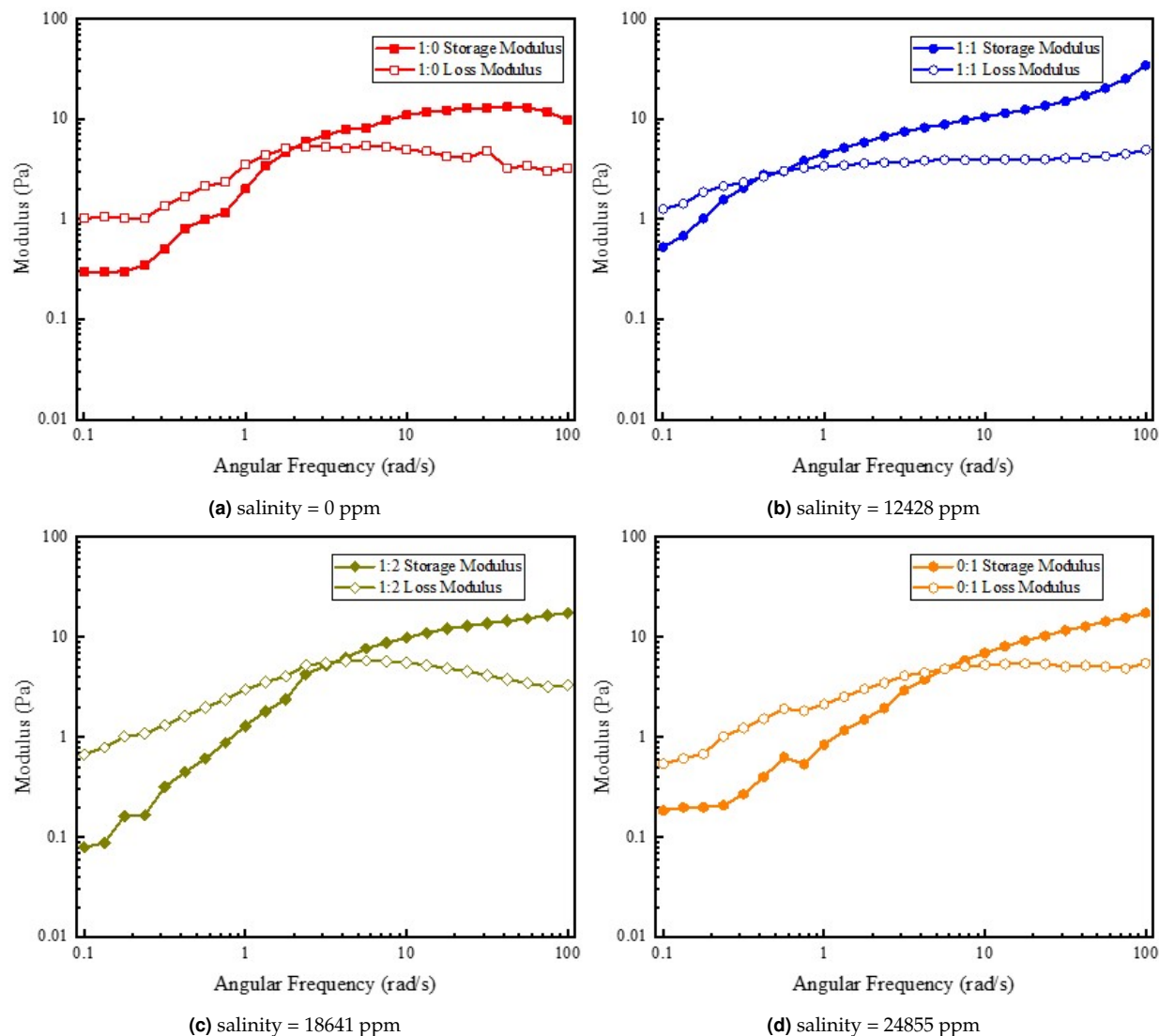


Fig. 7. Relationship between modulus and angular frequency of GACS solution.

Table 5. Viscoelastic characteristics of GACS solutions.

Salinity (ppm)	0	12428	18641	24855
$\tan \delta$	0.5375	0.3661	0.5361	0.6528
τ (s)	3.0784	7.1382	1.7749	1.1320

erations in the petroleum industry, particularly low permeability reservoirs. Based on this study, GACS exhibits outstanding properties that are desirable for enhancing oil recovery through surfactant flooding. The surface activity test showed that under different conditions of salinity, the CMC of GACS surfactant was in the range of 0.02 ~ 0.05 mmol/L. This indicated that at very low concentrations, the solution can self-assemble into micelle structures. GACS displayed low interfacial tension (10^{-2} mN/m) which significantly decreased to ultra-low values (10^{-4} mN/m) when

salinity increased to about 18600 ppm. Despite the decrease in viscosity of the GACS solution with an increase in salinity, the surfactant was capable to viscosify the solution even at the optimal salinity. Interestingly, GACS solutions displayed exceptional viscoelastic properties at different salinities which are more needed for EOR application. The study revealed that GACS can enhance oil recovery greatly by lowering oil-water interfacial tension to ultra-low values, improving the viscosity of displacing fluid, and imparting viscoelastic property for the solution. The viscosifying ability of GACS was found to be influenced by temperature change. However, the viscosifying effect was high enough to improve oil recovery under reservoir conditions (temperature and salinity).

5. Acknowledgements

This work was funded by the National Natural Science Foundation of China (51774049) and the National Natural Science Foundation of China (51474035).

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