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Development of a new surfactant-polymer system and its implementation in Dagang oilfield

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In this paper, a new surfactant-polymer enhanced oil recovery (EOR) system is investigated for the feasibility of injection in Dagang. The new SP flooding system has been designed and developed for Dagang oilfield with Dagang petroleum sulfonate (SLPS) as the primary ingredient. The dynamic behavior of the system and the interactions of the system components have been investigated through various methods, including dissipative particle dynamics (DPD) molecular modeling technology and dynamic interfacial-tension analysis. The results have shown a significant synergistic effect between sulfonate and nonionic surfactant. The interfacial tension (IFT) and its time to reach equilibrium could be dramatically decreased, suggesting a fast diffusion-adsorption characteristic of ionic surfactants as well as the high surface activity of nonionic surfactants. The SP flooding formulation was optimized. A pilot test has been carried out. The field trial provides useful information for the further large-scale application of the SP system in Dagang oilfield.

Key words: Surfactant-polymer flooding, sulfonate, interfacial tension, dynamic behavior, pilot field trial.

INTRODUCTION

After being developed for more than thirty years, Dagang oilfield becomes increasingly expensive to exploit due to high water cut in the main oilfield reservoir. Meanwhile, it is getting extremely difficult to explore new oil reservoirs. Therefore, it is of great importance to improve oil production of current oilfields by using new technologies.

Alkaline-surfactant-polymer (ASP) flooding invented in 1980s has been regarded as a potential enhanced-oil-recovery (EOR) technology which is more powerful than polymer flooding. Extensive studies on ASP technology have been carried out in the U.S., Germany, and the North Sea (Hernandez et al., 2003; Carrero et al., 2007; Yin et al., 2010). In China, a number of bench-scale and *on-site* experiments on ASP technology were carried out in Daqing and Dagang oilfields, which had indicated satisfactory capability of ASP systems to increase oil

production (Baoyu et al., 1994; Xulong et al., 2002; Kang, 2001). Meanwhile, disadvantages identified during the implementation process of the ASP flooding technology, such as, severe scaling in the injection lines and strong emulsification of the produced fluid, (Wang et al., 2005; 2009; Zhang and Xiao, 2007; Gao et al., 2010) also limited its further application in the field.

In order to overcome the drawbacks associated with ASP flooding, alkali-free surfactant-polymer (SP) flooding technology was developed, and its application in Dagang oilfield has been extensively investigated by authors' research institute. The pilot field trial of our S-P flooding system in southwest Dagang 7th region G-2 was the first field experiment of the S-P flooding technology in China. The objective of the work focused on demonstrating the feasibility of the S-P flooding technique for further

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Table 1. List of chemicals.

No.	Name	Company	Purity grade
1	Dodecyl polyoxyethylene polyoxypropylene ether C ₁₂ H ₂₅ (EO) ₄ (PO) ₅ H (LS ₄₅) and C ₁₂ H ₂₅ (EO) ₅ (PO) ₄ H (LS ₅₄)	Henkel company, Germany	Greater than 99.95%
2	Sodium hexadecyl sulfonate (AS);	Company, China	Purity grade
3	Sodium dodecyl sulfonate (SLS);		
4	Nonylphenol polyethylene oxide ether TX, n = 8 – 9		
5	Shengli petroleum sulfonate (SLPS)	Shengli zhongsheng	31.4% active

enhancement of oil recovery after polymer flooding. Moreover, the field trials provided useful information on the S–P flooding technology, such as, the formulation design and optimization. This could eventually lead to wider implementation of the S–P flooding system in Dagang oilfield.

Austad and Fjelde had previously indicated that significant improvements can be obtained by coinjecting surfactant and polymer at a rather low chemical concentration. Furthermore, the key factor in selecting chemicals is to avoid S–P complex formation in order to still maintain a very low IFT at low surfactant concentration (Austad et al., 1994; Gao et al., 2011). Among all the efforts to selecting good chemicals for S–P flooding, petroleum sulfonate, a good oil displacement agent, has gained more and more attention as used in chemical enhanced oil recovery technology. Zhang et al. (2004) studied the effect of different acidic fractions in crude oil on dynamic interfacial tensions in surfactant/alkali/model oil systems (Van der and Joos, 1980; Gao et al., 2009). A study by Al-Hashim had focused on the adsorption and precipitation behaviour of petroleum sulfonates on Saudi Arabian limestone (Zhang et al., 2004). DeBons and Whittington compared performance of the petroleum sulfonate with lignin in Berea sandstone cores (Al-Hashim et al., 1988; Gao et al., 2012). All these studies were limited on the laboratory development stage, none petroleum sulfonate–polymer pilot test have been report using Dagang petroleum sulfonate (SLPS) as the primary component, this effort studied the effect of the secondary surfactant and polymer in the formulation, as well as their capability to enhance the overall oil-recovery performance.

MATERIALS AND METHODS

Surfactant design for the S–P flooding system

Dodecylpolyoxyethylenepolyoxypropyleneether, C₁₂H₂₅(EO)₄(PO)₅H (LS₅₄), or C₁₂H₂₅(EO)₅(PO)₄H(LS₅₄), (both purchased from Henkel company, Germany) are colorless, viscous liquid with purity greater than 99.95%; sodium dodecyl benzene sulfonate (SDBS), sodium hexadecylsulfonate (AS), sodium dodecyl sulfonate (SLS), and nonylphenol polyethylene oxide ether (TX, n =8 to 9) are all of analytical purity grade, purchased from Shanghai Reagent

Company, China; Dagang petroleum sulfonate (SLPS) contains 31.4% active ingredient (Table 1).

Droplet volume method was utilized to measure dynamic surface tension; TX-500C spin drop apparatus from Bowing Industry Corporation, USA, was used to measure IFT. Viscosity of the polymers was examined using DVIII viscometer in reservoir conditions. Chromatographic separation tests were conducted for the surfactant flooding system under reservoir conditions. The tests were performed in a tube model with inner diameter of 1.5 cm and length of 50 cm. In the tests, unconsolidated model porous media composed of fine silica sands of different mesh was used and its permeability was about $1.5 \times 10^{-3} \mu\text{m}^2$ which was close to the reservoir conditions. The tube was prevacuumized and then saturated by water before injection of the surfactant flooding fluid of 0.3 PV. Afterwards, the tube was flooded by water until the outgoing concentration of surfactants became zero.

In oil displacement test, the formation water and re-injection water were prepared at salinity of 4876 and 6188 mg/L, respectively. The oil mixture was formulated by kerosene and dehydrated crude oil from Dagang 16 to 011 well to simulate underground crude oil at 50 mPa.s viscosity. Testing temperature was 70 °C. The tests were performed in a core with inner diameter of 2.5 cm and length of 30 cm. The heterogeneousness of the formation was simulated by dual-core system of different permeability: 1500×10^{-3} and $4500 \times 10^{-3} \mu\text{m}^2$. The core-flooding procedure is as follow:

Vacuum-pump test core for 2 h, then saturated with reservoir water. Afterwards the core was flushed with (1) oil until a state corresponding to S_{wi} was reached (2) water until a state corresponding to S_{or} was reached and (3) a slug of surfactant formula until water cut to 100%. Injection speed was 0.23 mL/min.

RESULTS AND DISCUSSION

Dynamic surface tension of the sulfonate–nonionic surfactant system

SLPS was been selected as the primary surfactant of the S–P flooding system for the pilot field trial in Dagang oilfield due to its compatibility with oil reservoir as SLPS is produced directly from Dagang crude oil. Meanwhile Employing SLPS as the main component of the S–P flooding formula also lowers the reliance on the outside chemical sources. As an anionic surfactant, SLPS shows strong electric repulsion among the polar heads. A variety of hydrophobic chains structures is expected as SLPS is produced from Dagang crude oil. This results in a loose

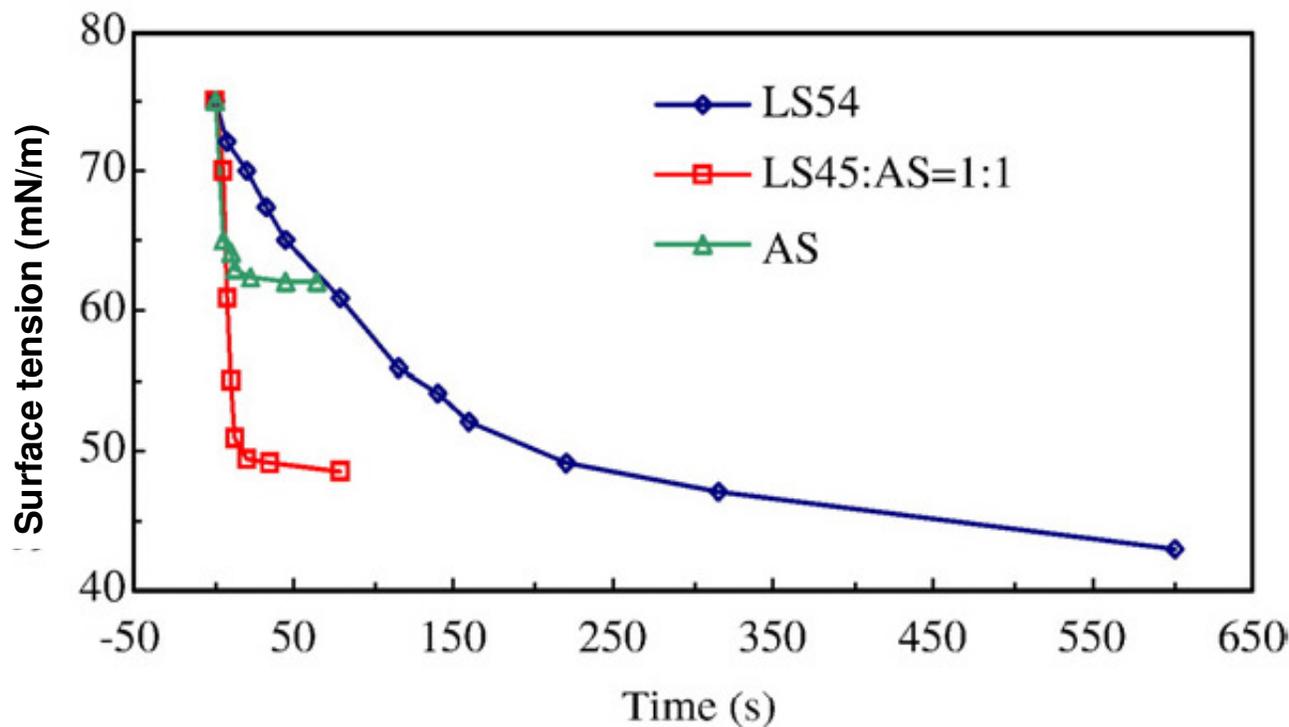


Figure 1. Dynamic surface tension of the sulfonate–nonionic surfactant system.

arrangement of chains in the interfacial membrane and thus the low interfacial activity. As a consequence, SLPS itself is incapable of decreasing the oil/water IFT to an extremely low level of 10^{-3} mN/m. Nevertheless, previous studies had indicated that co-adsorption of different types of surfactants in oil/water interface could generate interfacial membranes with tight and ordered arrangement of molecules due to weaker steric and electric interactions in the system (Salager and Mongan, 1979; Myers, 2009; Sheng and Wang, 2001; Gao et al., 2010). In this case, the oil/water IFT can be further reduced as a result of the synergistic effects between different surfactants.

The dynamic surface tension of AS-LS54 was measured, as shown in Figure 1, to investigate the activating behavior of the sulfonate–nonionic surfactant system and the synergistic interactions between the two types of surfactants.

Figure 1 illustrates that the surface tension for AS equilibrates immediately in water while it takes a much longer time for LS54. The equilibrium surface tension are determined to be 62 mN/m for AS and 42 mN/m for LS54, respectively, which is 20 mN/m lower than that for AS. In contrast, the surface tension of AS-LS54 (in 1:1 ratio) reaches the steady state as efficient as AS, and possesses a low value similar to that for LS54. Since AS-LS54 shows augmentation of diffusion coefficient as well as surface activity compared to the single surfactant system, sulfonate–nonionic surfactant has been

determined to be the basic formulation of the S–P flooding system (Zhao et al, 2010).

Synergistic effect between sulfonate and nonionic surfactant

Structure–function relationships have been investigated for combinations of sulfonate and various nonionic surfactants through measurement of oil/water IFT. Total concentration of each surfactant system studied is 0.05% by weight in 0.7% NaCl brine.

Table 2 demonstrates a remarkable decrease of 1-octane/water IFT upon the addition of a small amount of nonionic surfactant (TX-100 or Tween-80) in SDBS, suggesting a certain synergistic effect between SDBS and nonionic surfactants. With toluene as the oil phase, combination of SDBS and Tween gives the lowest oil/water IFT among all the surfactant systems. It has been found that the oil/water IFT gradually increases with the content of saturated alkanes in oil phase. Lower oil/water IFT by introducing toluene into saturated alkanes shows that structure similarity between surfactant hydrophobic chains and oil phase molecules favors the IFT reduction. Clearly, the most prominent synergistic effect for SDBS comes from the combination of nonionic surfactants that contain aromatic rings in the hydrophobic chains. Since the ratio of aromatic hydrocarbon and alkane in Dagang crude oil is near 1:2

Table 2. IFT (mN/m) for sulfonate and sulfonate–nonionic surfactant systems.

Oil phase surfactant							
Oil	SDBS	SDBS: LS45=9:1	SDBS:TX- 100=8:2	SDBS:TW- 80=8:2	SDBS: AES=9:1	SLS	TX
1-Octane	1.209	/	0.528	0.983	/	>3	0.5
Toluene:1- Octane=1:2	0.456	0.505	0.142	0.321	0.418	2.46	/
Toluene:	0.333	/	0.227	0.082	/	/	>3

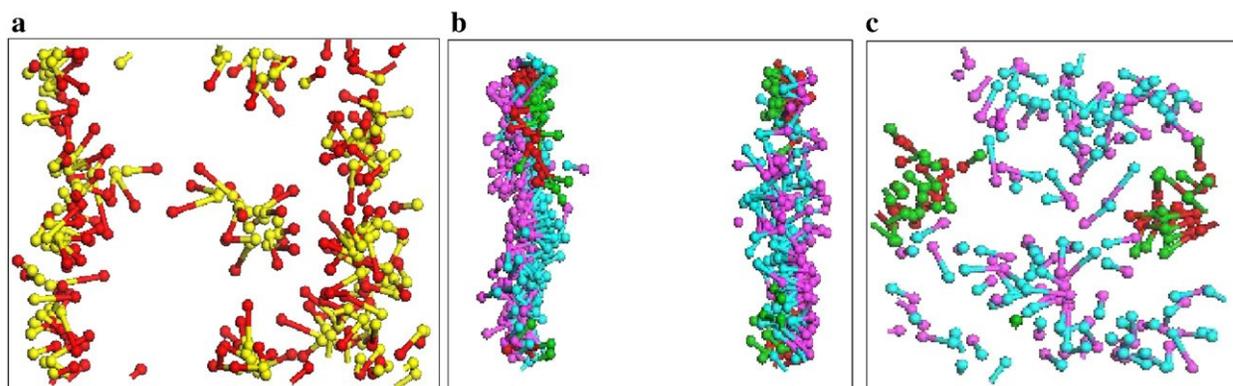


Figure 2. Arrangement of sulfonate and sulfonate–nonionic surfactant in the interface through molecular modeling. (a) SDBS array in the interface. Red and yellow colors represent the head and tail of SDBS respectively. (b), (c) SDBS-TX array in the interface. Pink and blue colors represent the head and tail of SDBS respectively, while red and green colors correspond to the head and tail of TX respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

the combination of the right molecule chain and the number of EO with SLPS gain the lowest interfacial tension in surfactant–polymer flooding system.

Molecular modeling of the synergistic effect between sulfonate and nonionic surfactants

Dissipative particle dynamics (DPD), one of the molecular modeling techniques, was used in this study to simulate the synergistic effect between sulfonate and nonionic surfactants (Dong et al., 2004). DPD originally proposed by Hoogerbrugge and Koelman in 1992 is a state-of-the-art mesoscale simulation method for the study of complex fluids, such as polymeric or colloidal suspensions. In DPD, molecular cluster in complex fluid system is denoted as ‘bead’, which is taken to be an effective soft sphere that acts as a center of mass. Based on Newton’s motion equation, each bead interacts with the remaining beads through soft potentials, subjected to dissipative and fluctuating forces. In current simulation, each surfactant molecule was represented by two beads, hydrophilic head bead and hydrophobic tail bead, linked by elastic springs with elastic constant to be 4.0 KT.

Simulation was carried out in a $20 \times 10 \times 10$ simulation box at density of 3.0 using 20,000 time steps and a time step interval of 0.05. The mass of bead and system temperature were all set to be 1.0 DPD unit.

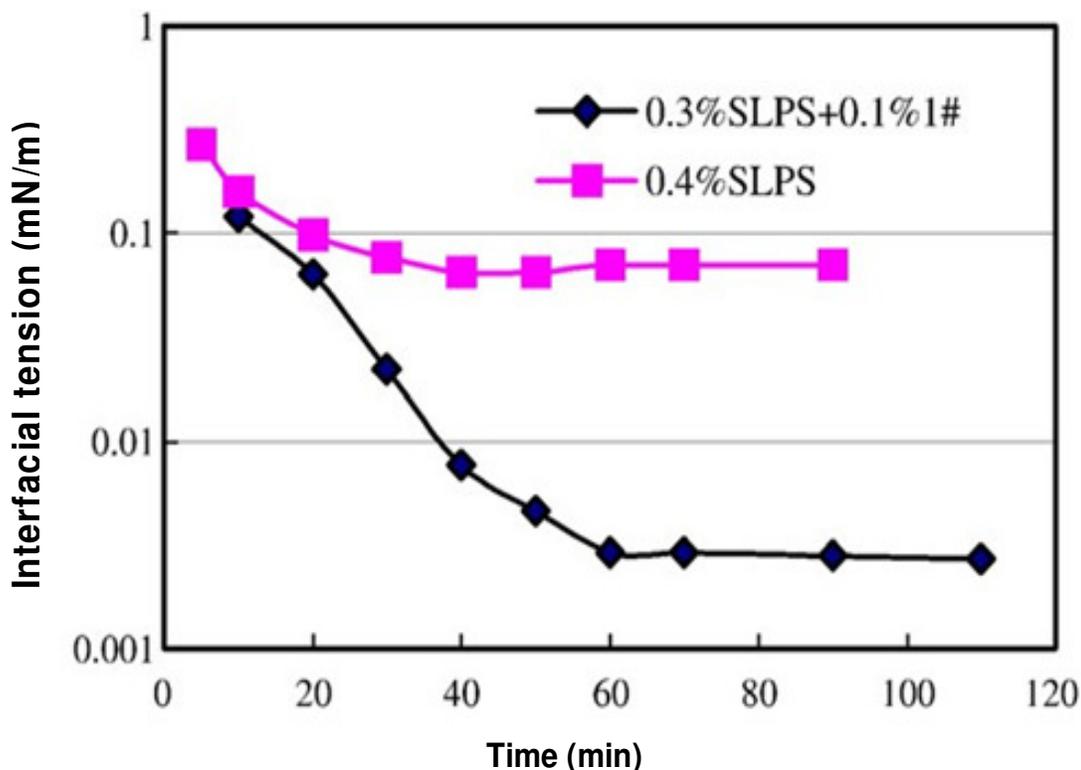
Simulation (Figure 2a) shows loose arrangement of SDBS in the interface with cavities that can not be inserted by other free SDBS molecules no matter how large the concentration of SDBS is. However, the TX clusters can enter the cavities (Figure 2b, c) because of the weaker repulsion between the nonelectric polar head of TX and electric polar head of SDBS than that between two electric polar heads of SDBS. Therefore, combination of TX with SDBS offers a synergistic effect to tremendously diminish the IFT by significantly increasing the surfactant density in the interface.

Surfactant formulation design for the S–P flooding system in Dagang oilfield

The crude viscosity was determined to be 45 mPa.s and the reservoir temperature to be 68°C in the southwest Dagang 7th oilfield Ng54-61. The estimated salinity is 6188 mg/L for injection water and 8207 mg/L for produced

Table 3. IFT of SLPS and its combination with various surfactants.

Number	Surfactant system	IFT (mN/m)
1	0.3%SLPS	7.62×10^{-2}
2	0.3%SLPS-01+0.1%JDQ-1	8.61×10^{-3}
3	0.3%SLPS-01+0.1%JDQ-2	5.65×10^{-3}
4	0.3%SLPS-01+0.1%1#	2.95×10^{-3}
5	0.3%SLPS-01+0.1%4#	6.03×10^{-3}
6	0.3%SLPS-01+0.1%T1501	9.81×10^{-3}
7	0.3%SLPS-01+0.1%T1402	6.00×10^{-3}
8	0.3%SLPS-01+0.1%4-02	5.10×10^{-3}

**Figure 3.** Dynamic IFT of the surfactant flooding system.

water. The bivalent ion (Ca^{2+} and Mg^{2+}) in injection water is 189 mg/L.

Based on aforementioned synergistic studies and reservoir conditions, SLPS was formulated as the primary ingredient together with a variety of complementary surfactants of different types and structures. Table 3 shows the oil/water IFT for these formulations. The formulation with the lowest IFT (2.95×10^{-3} mN/m) in Table 3 corresponds to the combination of SLPS and the secondary surfactant, 1#, which is a nonionic surfactant with TX-100 as the basic ingredient. Furthermore, the dynamic oil/water IFT for both SLPS and 1# has been illustrated in Figure 3.

Chromatographic separation of surfactants in the S–P flooding system in Dagang oilfield

An important consideration of S–P flooding system is to avoid possible chromatographic separation of surfactants, which occurs during the movement of the flooding chemicals in the oilfield formation since the flooding system is composed of surfactants of various structures. Undoubtedly, chromatographic separation would dramatically decrease the flooding efficacy and oil recovery (Austad et al., 1994; Wang et al., 2005; Sui Xihua et al., 2000).

The dynamic adsorption of the injected SLPS-1#

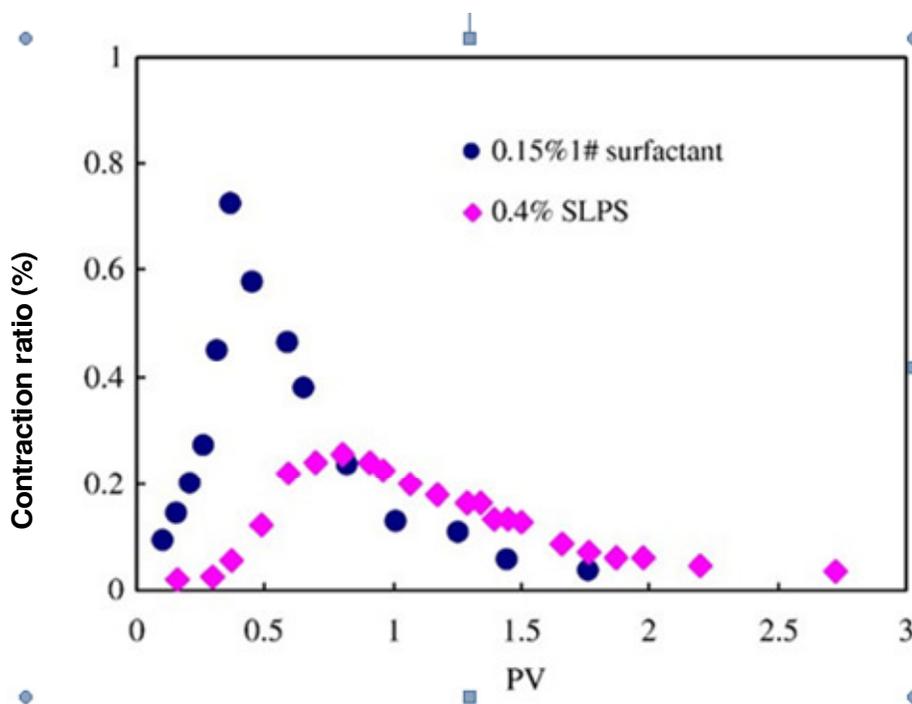


Figure 4. Dynamic adsorption of SLPS and 1#.

mixture in Figure 4 suggests that there exists a chromatographic separation phenomenon between SLPS and 1#. The time difference of 0.5 PV between the outgoing concentration peaks of these two surfactants was observed. To decrease the possible surfactant adsorption and chromatographic separation in the oilfield formation, it has been suggested to increase the total concentration of the injected surfactants to be higher than the threshold value of 0.15%. As noted before, the IFT for SLPS-1# is able to achieve the super low level (10^{-3} mN/m) as long as the total concentration of two surfactants is above 0.15% and below 0.65%. Synergism between SLPS and 1# might be weakened at the front edge of the flooding fluid because of the dilution effect by underground water and the adsorption of surfactants in the formation. Based on above studies, it is recommended to start with 0.45% SLPS + 0.15% 1# in the field trial.

Polymer design for the S–P flooding system

Polymer viscosity swept volume by polymer (Cao et al., 2002). As a result, it is recommended to incorporate polymer into the surfactant flooding system to maximize oil recovery. Four polymers have been carefully selected for the S–P flooding system based on the oil recovery effects in the previous field applications. Viscosity of the polymers was examined using DVIII viscometer in reservoir conditions, that is, brine salinity of 6188 mg/L

and temperature of 68°C. The experiment shows high viscosity for all of the four polymers at concentration of 1500 mg/L, as shown in Figure 5.

Influence of polymer on the interfacial tension

Various polymers (0.15%) have been added into the surfactant flooding system consisting of 0.3% SLPS and 0.1% 1#, and their influence on the IFT has been evaluated accordingly. Because of the elevation of system viscosity upon the addition of polymers, the diffusion of surfactant from water phase towards oil/water interface slows down, extending the time for IFT to reach the super low level (Figure 6). Nevertheless, there is no difference on the order of the lowest IFT for both the S–P flooding system and polymer-free surfactant system, indicating that addition of polymer does not affect surfactants' ability to reduce the oil/water IFT.

Oil displacement test

Oil displacement tests were performed to investigate the EOR performance of different S–P flooding systems under reservoir conditions: actual formation temperature, pressure, permeability, and the degree of oil saturation. The results are shown in Table 4.

It has been found that an oil recovery enhancement of

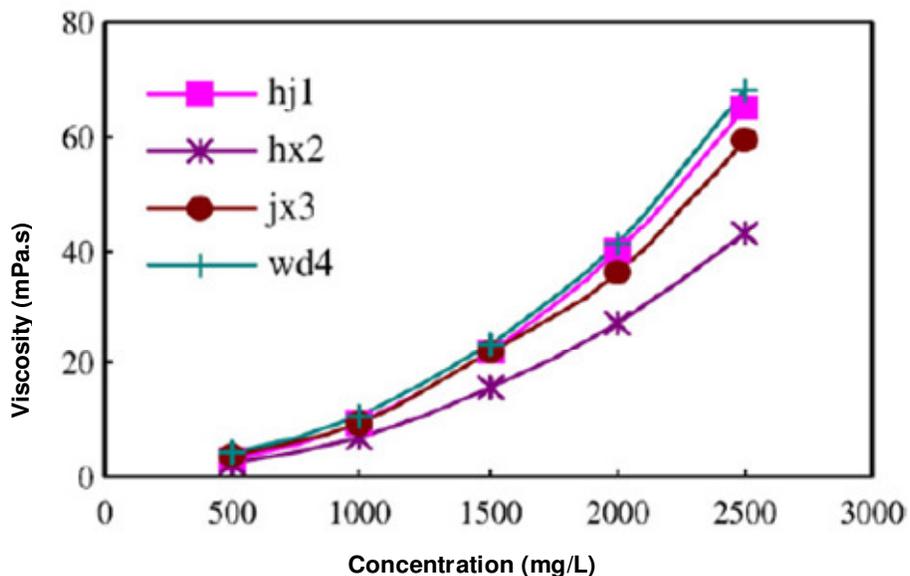


Figure 5. Polymer viscosity-concentration relationship.

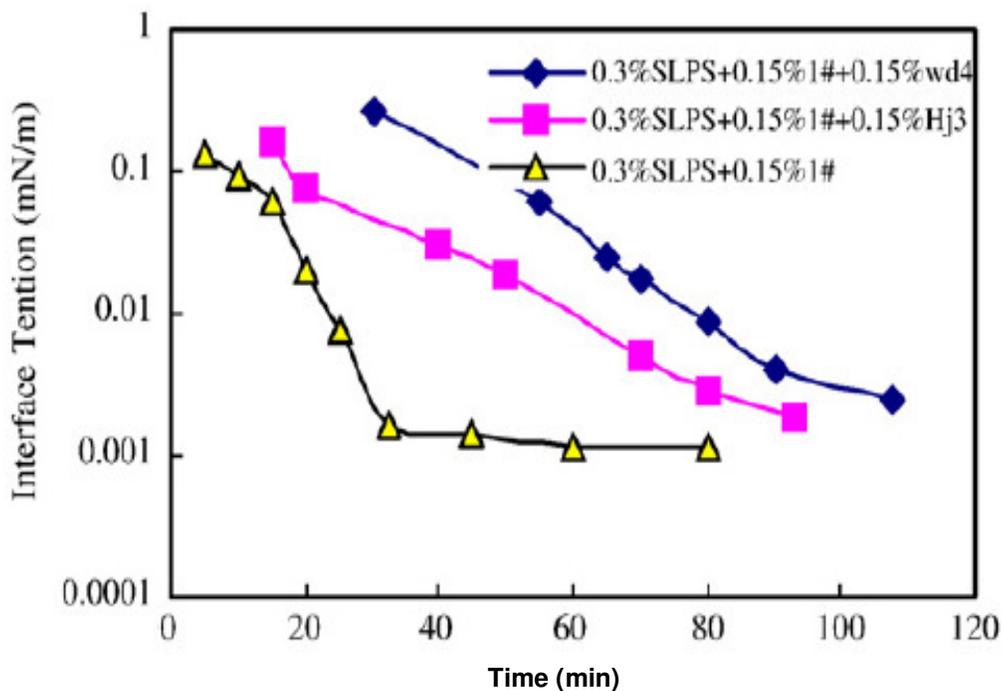


Figure 6. Influence of polymer on the IFT of surfactants in the flooding system.

Table 4. EOR comparison between S-P flooding and polymer flooding.

No.	Formulation	Injection (PV) (%)	OOIP %
Model-11#	0.3%SLPS+0.1%1#+0.15%P	0.3	18.1
Model-18#	0.15%P	0.54	15.2
Model-6#	0.15%P	0.3	11.7

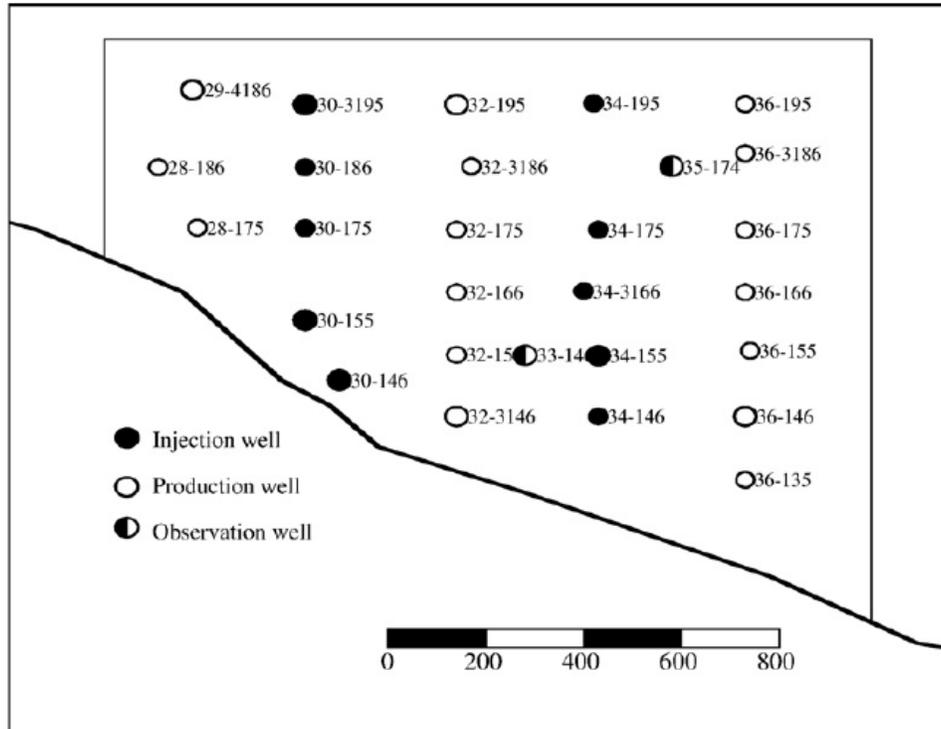


Figure 7. Well distribution for the pilot field trial of the S-P flooding system in G-2 oilfield.

18.1% can be achieved by injecting 0.3% SLPS + 0.1% 1# + 0.15% hj1 of 0.3 PV. Tests also showed that S-P flood outperformed polymer flood under the same core conditions and economical cost. Considering the adsorption consumption and costs, the polymer concentration is suggested to be 0.15 to 0.2% in the S-P flooding system.

Application of the S-P flooding system in the pilot field trial in Dagang oilfield

The pilot experiment of the S-P flooding system was carried out in the southeast zone of west Dagang 7th oilfield Ng54-61, which is characteristic of oil containing area of 0.94 km², and oil reservoir of 277.5 × 10⁴ t in depth of 1261~1294 m. The target zone consists of three oil containing strata (54, 55, and 61) with approximately 34% porosity.

The field trial involved twenty six wells, including sixteen production wells, ten injection wells, and three observation wells, as shown in Figure 7. The water cut of the field was determined to be 98.2%, and the oil recovery to be 34.4% before the trial. In order to alleviate “fingering” and “crossflow” of the flooding system in the formation, the three-slug injection methodology was established in the field trial. The first slug was the polymer pre-protection slug of 2000 mg/L polymer solution of 0.05 PV. The second slug was the main slug

of 0.3 PV solution consisting of 1700 mg/L polymer, 0.45% SLPS, and 0.15%1# and the third one was the polymer post-protection slug of 1500 mg/L polymer solution of 0.05 PV.

The pilot field trial has showed significant water-cut reduction and oil- production enhancement since the injection of the main slug of the S-P flooding system in June 2004. Most current field data show that the water cut has been continuously decreased by 13%, that is, from 98.2% in year 2004 to 85.2% in year 2007. Until July 2008, the oil production had increased dramatically by 159 t/day, that is, from 34 t/day to 193 t/day. The single well's oil production has reached to 2.0 × 10⁴ t and the cumulative oil production had risen by 11.5 × 10⁴ t with the oil recovery enhancement of 4.15%. Fourteen out of sixteen production wells in the field trial have manifested water-cut reduction and oil-production improvement at various degrees. The increase rates from field trials are clearly higher than that of exclusive polymer flooding.

Conclusion

The SP formulation in this work exhibits the efficient diffusion- adsorption properties of ionic surfactants as well as the high surface activity of nonionic surfactants.

An excellent synergistic effect has been obtained between the primary surfactant (SLPS) and secondary nonionic surfactant.

The finalized S–P flooding formulation used in the pilot field trial is capable to reduce the IFT to 2.95×10^{-3} mN/m, and improve the oil recovery by 18.1% in laboratory oil displacement tests. Since the injection of the main slug of the S–P flooding system in June 2004, the field trial has demonstrated tremendous decrease of water cut and enhancement of oil production. It has been reported that the accumulative oil-production increase had reached 17.8×10^4 t in July 2008. The increase rate of oil recovery and decrease rate of water cut from S–P flooding are clearly higher than those using single polymer flooding.

Current studies of surfactant dynamic activities and synergistic effects for the S–P flooding system will provide theoretical guidance for the future design and development of combination flooding systems. Meanwhile, the success of the pilot field trial of the S–P flooding system in Dagang oilfield will build up a solid foundation for the further large-scale application of the system.

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