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Research on the Synthesis and Application of AminoSulfonic Acid Gemini Surfactant

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ABSTRACT

Oil is a non-renewable resource, and it is essential to improve oil recovery. Surfactant flooding is a key technology in enhancing oil recovery. The present work aims to develop a new surfactant to the combination flooding system in oil displacement. The Aminosulfonic Acid Gemini Surfactant was prepared by using Hexadecylamine, 1-Butanaminium,N,N,N-tributyl-,bromide (1:1) and Sodium 2-chloroethyl sulfonate. The AminoSulfonic Acid Gemini Surfactant Sodium N,N'-Dihexadecyl-2-propanol- 1,3-ethylsulfonate SNNDPE) has better reaction temperature at 80°C. The best reaction time of SNNDPE is 12 h. Next, We made a characterization of SNNDPE. A new gemini surfactant was developed, which is beneficial to the research of material science. Then the performance of the new Gemini surfactant was tested. The surface tension tests suggested that the newly synthesized Aminosulfonic Acid Gemini Surfactant has higher surface activity. The resistance temperature of the Aminosulfonic Acid Gemini Surfactant solution is 80°C. Moreover, the oil-water interfacial tension experiment suggested that the minimum aqueous solution of the combination flooding system reaches an ultra-low level of 9.4×10^{-4} mN/m. The combination flooding system experiment results can be considered as a guidance in the design and studies of the combination flooding system for a selective.

Keywords: Gemini surfactant, Surface tension, Combination flooding system, Recovery.

INTRODUCTION

Oil is a non-renewable resource, and it is essential to improve oil recovery [1]. In practice, once the water content of the oil field is too high, water flooding cannot meet the actual production needs, and the remaining oil and residual oil in the reservoir cannot be recovered smoothly, which greatly reduces the oil recovery efficiency. A chemical flooding method can significantly improve oil recovery by changing the performance of oil reservoirs in rock pores. Surfactant flooding is a key technology in enhancing oil recovery, and it is the most stable oil displacement method of chemical flooding, which mainly cleans the residual oil and residual oil in the pores of the reservoir to improve oil recovery [2]. Surfactant have good properties of reducing surface tension [3], penetration [4], solubilization [5] and dispersion [6]. And surfactant is widely used as oil-displacing agents [7]. In the 1970s, Gemini surfactant was developed as a new type of surfactant [8]. Gemini surfactant molecular was composed of two linked monomer [9]. Gemini surfactant had lower interfacial tension and critical micelle concentration than traditional surfactant. Gemini surfactant also showed outstanding characteristics in water solubility, solubilization, and viscoelasticity. Amphoteric Gemini surfactant had shown good synergistic effects in emulsification, biodegradability, and dispersibility, which had attracted the attention of a large number of academic researchers and domain experts [10–11].

Zhanfeng Xie *et al*. developed series of alkylbetaine zwitterionic gemini surfactants. The experiment showed that this series of surfactants has a lower critical micelle concentration (CMC) and a strong ability to reduce the interfacial tension of the solution. The logCMC of C_nA_b decreased linearly with increasing when the chain lengths up to 12. Viscosity measurements showed only $C_{10}A_b$ could increase the viscosity of high-concentration aqueous solutions [12]. Peiqian Li *et al.* synthesized a new type of didodecylmethylcarboxyl betaine ($diC_{12}B$) to reduce the amount of alkaline substances. The experiment showed that when the mass concentration was about 0.01%–0.5%, the interfacial tension of the Gemini surfactant reached 10^{-3} mN/m at 45°C.

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When mixed with other hydrophobic and hydrophilic sulfobetaines in surfactant-polymer (SP) flooding free of alkali, the $diC_{12}B$ gives a high recovery. Moreover, the $diC_{12}B$ reduce the amount of alkaline substances (as the oil-displacing agent component) [13]. Many experts want to improve the micelle capacity of surfactants. Two tetrasiloxane Gemini imidazolium surfactants with methylene spacer groups ($[Si4\text{-}s\text{-}Si4im]Cl_2$, s = 4, 6) were synthesized by Xiaohui Zhao *et al.* A series of surface activity parameters (cmc, γcmc, πcmc, pc20, Γmax and Amin) and the adsorption isotherms were obtained from the surface tension plots. The results show that the tetrasiloxane Gemini imidazolium surfactant has the higher capacity to form micelles [14]. Yancheng Zheng *et al.* synthesized 1,2-bis[N-methyl-N-(3-sulfopropyl)- alkylammonium]-ethane betaine (GCS_{12}), which is highly sensitive to salt. The CMC value of GCS_{12} is 0.07 mmol/L in distilled water. By measuring the CMC of GCS_{12} and the nonionic surfactant lauric acid diethanolamide (CDA) at different molar ratios, the interaction between GCS_{12} and CDA was studied.[15]. Hongqin Liu *et al.* synthesized a series of amine-oxide Gemini surfactants featuring amide groups to reduce the surface tension of water. The surface activities of surfactants have been measured. The results indicate that amine-oxide Gemini surfactants reduced the surface tension of water to a minimum of about 26.91 mN/m at a concentration of 2.92×10^{-5} mol/L [16].

Alkaline flooding exists in composite flooding nowadays. It is very important to find an alkali-free composite oil-displacing agent that achieve ultra-low interfacial tension [17]. Surfactant and alkaline chemicals are usually used to reduce the capillary force during chemical flooding. It is well known that the surfactant molecules can accumulate at the oil/water interface to reduce the oil/water interfacial tension (IFT). The alkali-free oil-displacing agent can not only save costs, but also prevent alkali scale [18]. Amphoteric Gemini surfactant is the current research frontier [19]. In this paper, a new type of Aminosulfonic Acid Gemini Surfactant was synthesized. The AminoSulfonic Acid Gemini Surfactant Sodium N,N'-Dihexadecyl-2-propanol-1,3- ethylsulfonate SNNDPE) has better reaction temperature at 80°C. The best reaction time of SNNDPE is 12 h. We tested the oil-water interfacial tension and combination flooding system of the AminoSulfonic Acid Gemini Surfactant. Then, the combination flooding system was carried out on the AminoSulfonic Acid Gemini Surfactant.

2. EXPERIMENTAL PROCESS

2.1. Materials and apparatus

Materials: All chemicals used for synthesis were analytical grade without further purification. The chemical reagents used such as 1-Butanaminium,N,N,N-tributyl-, bromide(1:1)(TBAB), Methanol(CH_4O), Ethanol(C_2H_6O), 2-Propanone(C_3H_6O), sodium hydroxide(NaOH), Sodium carbonate($NaCO_3$), 1-Hexadecylamine($C_{16}H_{35}N$), Sodium 2-chloroethyl sulfonate($C_2H_6ClNaO_4S$), 1,3-Dichloro-2-propanol($C_3H_6Cl_2O$) and propan-2-ol(C_3H_8O) are from Sinopharm Purchased by Chemical Reagent Co., Ltd. Distilled water was used for all experiments.The oil sands used in the experiment were from Daqing oil sands.

Apparatus: ALC-210.4 electronic analytical balance (Sartorius AG, Germany); 2XZ-4 type rotary vane vacuum pump (Wenling Suli Motor Factory); DK-98-IIA electric heating constant temperature water bath (Tianjin Test Co., Ltd.); GZX-9023MBE digital display blast drying oven (Shanghai Boxun Industrial Co., Ltd. Medical Equipment Factory); AWJ-Y Constant Speed Stirrer (Jintan Hongke Instrument Factory, Jiangsu Province); ZL3000 Automatic Interface Tension Tester (Zibo Hainuo Instrument Factory); Precision Pressure Sensor (Xi'an High Precision Instrument Factory); DYQ-1 type multifunctional core displacement device (Haian County Petroleum Scientific Research Instrument Factory).

2.2. Experiment of synthesis and performance

The Aminosulfonic acid Gemini Surfactant was prepared by using Hexadecylamine, 1-Butanaminium,N,N,N-tributyl-, bromide(1:1) and Sodium 2-chloroethyl sulfonate. Then, we studied on influence of reaction temperature and reaction time on the synthesis of SNNDPE. Last, we made a characterization of SNNDPE. About performance testing, we made a resistance temperature of the Aminosulfonic Acid Gemini Surfactant solution. Then, we tested surface tension and oil-water interfacial tension of the Aminosulfonic Acid Gemini Surfactant solution. In the end, we made the combination flooding system of Aminosulfonic Acid Gemini Surfactant.

3. SYNTHESIS AND DISCUSSION

3.1. Synthesis of Aminosulfonic Acid Gemini Surfactant

Clean the 500 mL three-necked reaction flask with distilled water. Put a thermometer, a constant pressure dropping funnel and a five-chamber reflux condenser into the three openings of the three-necked reaction flask.

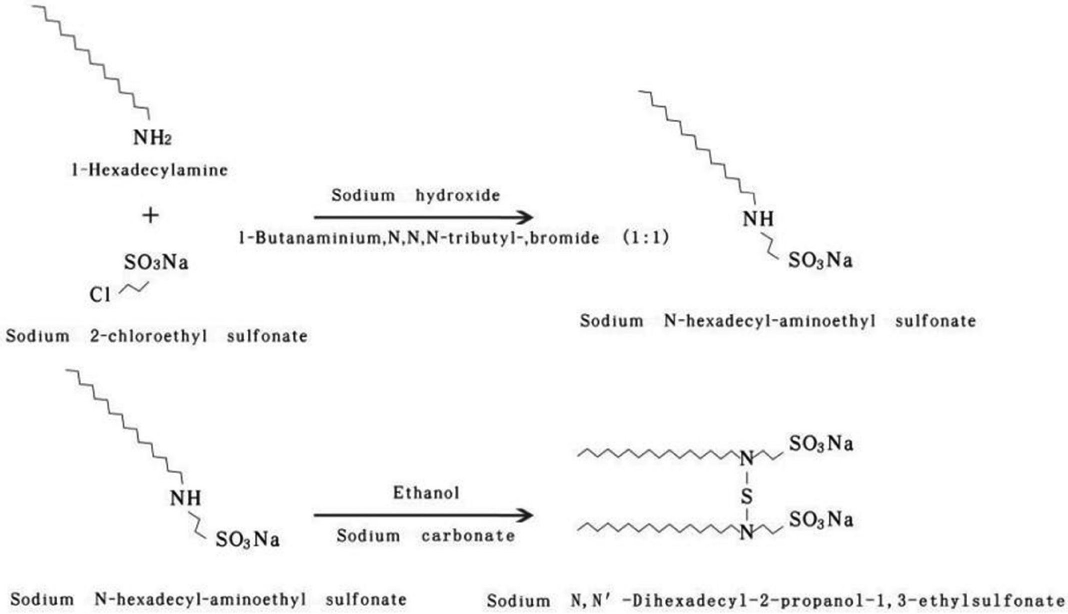

Figure 1: The reaction mechanism of SNNDPE.

Add 250 ml of C_3H_8O and 0.2 mol of $C_{16}H_{35}N$ to the reaction flask. Then add the 5 mol/L NaOH aqueous solution to the reaction flask and keep the pH 8.5. Add 0.001 mol of TBAB as a catalyst in the reaction flask. The temperature of the reaction liquid is adjusted to 50°C. Add 0.18 mol of $C_2H_6ClNaO_4S$ at 50°Cand increase the Solution temperature to 85°C to react for 6 hours. After the reaction, the solution is cooled to room temperature. Then the solvent is removed by rotary evaporation. The obtained product is recrystallized with ethanol. Obtain intermediate Sodium N-hexadecyl-aminoethyl sulfonate(SNHAS) by Suction filtration and drying in a drying oven (Figure 1).

Clean the 500 mL three-necked reaction flask with distilled water. Put a thermometer, a constant pressure dropping funnel and a five-chamber reflux condenser into the three openings of the three-necked reaction flask. Then add the rotor to the reaction flask. Put 0.15 mol of SNHAS, 200 mL of ethanol and 0.2 mol of $NaCO_3$ into the three-necked reaction flask. The temperature of the reaction flask is adjusted to 70°C. Add 0.08 mol of $C_3H_6Cl_2O$ to 80 mL of Ethanol solution, then add it to the reaction solution, and start the reaction for 12 hours. The reaction liquid is cooled and filtered with suction when the reaction was over. Wash with acetone, wash impurity, mix methanol and acetone, and then subject to recrystallization. Finally, it is dried in a drying oven to get the AminoSulfonic Acid Gemini Surfactant Sodium N,N'-Dihexadecyl-2-propanol-1,3-ethylsulfonate (SNNDPE) (Figure 1).

3.2. The influence of reaction temperature on the synthesis of SNNDPE

The reaction was carried out at the temperatures of 70°C, 75°C, 80°C, 85°C and 90°C. The interfacial tension (Figure 2) of the product was tested [20].

The figure showed that the interfacial tension decreased gradually with the increase of temperature, and the interfacial tension reached the minimum value when the temperature reached 80°C. After 80 °C, the peak value of the interfacial tension increases gradually with the increase of temperature. According to the experimental data, the optimal reaction temperature is 80°C. Therefore, 80°C was selected for experimental operation and synthesis reaction.

3.3. The effect of reaction time on the synthesis of SNNDPE

The reaction was carried out at the reaction times of 10 h, 11 h, 12 h, 13 h and 14 h. The product was carried out by interfacial tension test (Figure 3).

As shown in Figure 3, the peak of interfacial tension decreased with the increase of time before 12 h. The minimum reached at 12 h. After 12 h, it is obvious that the peak of interfacial tension became larger with the increase of time. According to the experimental data, the best reaction time is 12 h. Therefore, The 12 h was chosen for experimental operation and synthesis reaction.

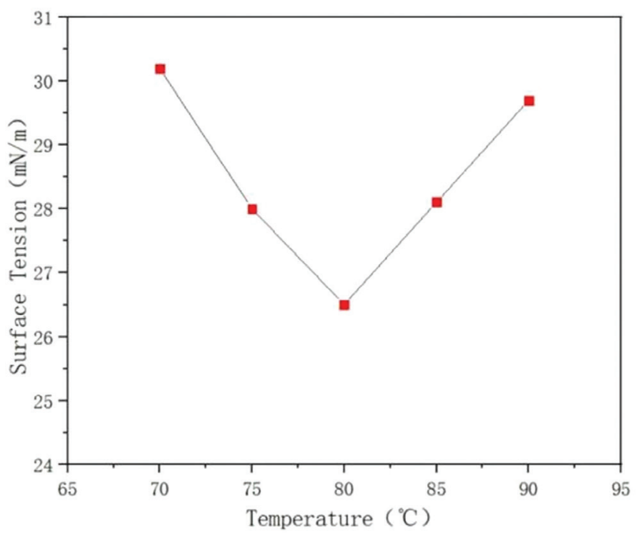

Figure 2: The relationship between temperature and interfacial tension.

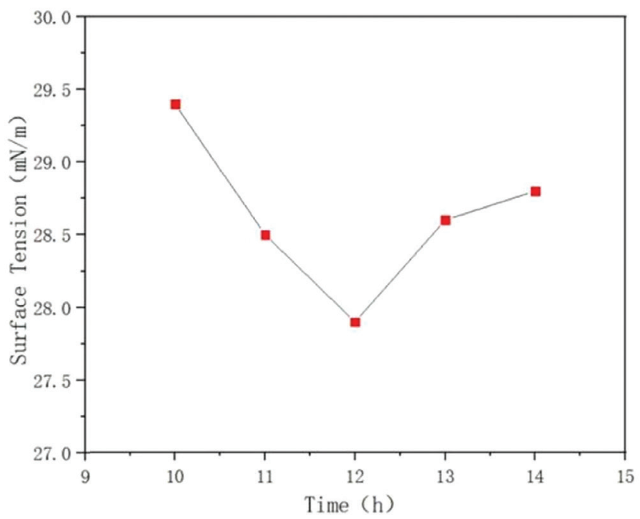

Figure 3: Graph of the relationship between reaction time and interfacial tension.

3.4. Chemical characterization of SNNDPE

The experimental result was characterized by infrared spectroscopy (Figure 4). The characterization result of the experimental samples showed that the SNNDPE contains sulfonic acid groups in the molecular structure. The middle gasket are ether group and hydroxyl group.

The C-H symmetrical stretching vibration peaks of methyl and methylene are at 2919.24 cm^{-1} and 2850.75 cm^{-1} respectively, and the asymmetrical stretching vibration peaks of methyl is at 1441.21 cm^{-1}. The characteristic vibration peak of the sulfonic acid group are at 1200.88 cm^{-1}, 1050.68 cm^{-1}, 617.81 cm^{-1} and 529.23 cm^{-1}. In addition, the stretching vibration absorption band of -OH is at 3422.93 cm^{-1}, at 1050.68 cm^{-1} is peak of the CN stretching vibration absorption. It is obvious that the SNNDPE has been synthesized successfully.

4. PERFORMANCE TESTING AND DISCUSSION

4.1. The resistance temperature of the Aminosulfonic Acid Gemini Surfactant solution

The Aminosulfonic Acid Gemini Surfactant solution was weighed 0.1 mol. In order to investigate the temperature resistance of the Aminosulfonic Acid Gemini Surfactant (Figure 5), the Aminosulfonic Acid Gemini Surfactant solution placed at 60°C, 70°C, 80°C and 90°C by the two-phase titration method (The principle of the two-phase titration method is to use the property of benzoic acid to dissolve in organic solvents, add an organic

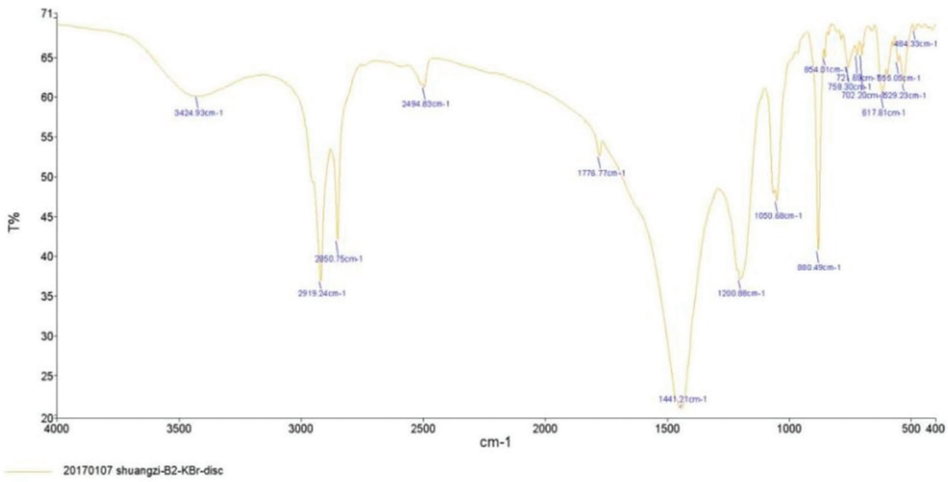

Figure 4: Infrared Spectra of SNNDPE.

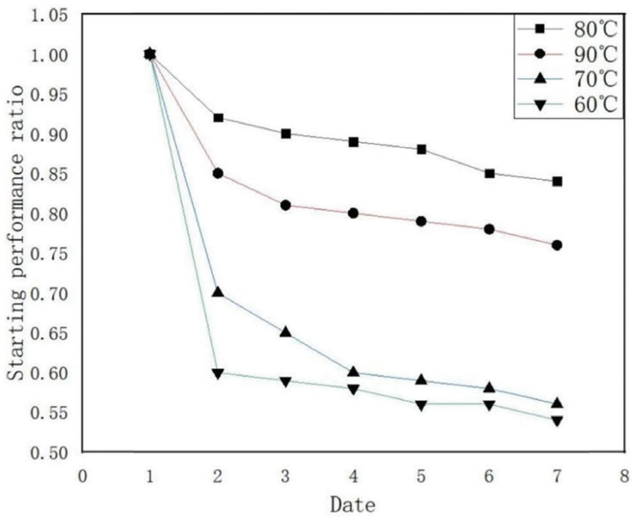

Figure 5: Temperature resistance of Aminosulfonic Acid Gemini Surfactant solution.

Table 1: The relationship between the interfacial tension and concentration of Aminosulfonic Acid Gemini Surfactant.

SOLUTION CONCENTRATION (mmol/L)	0.01	0.02	0.03	0.04	0.05	0.1	1.5	2.5	3.5	5
SURFACE TENSION (mN/m)	59.5	50.4	42.2	32.1	25.7	24.8	24.2	24.0	23.9	23.7

solvent that is immiscible with water in the water phase, and continuously extract the generated benzoic acid into the organic phase during the titration process to reduce the amount of benzene. The concentration of formic acid in the aqueous phase and the dissociation of benzoic acid are reduced, so that the titration reaction is completed and the end point is clear) [21]. The concentration of the solution was determined.

The vertical axis is the concentration of the solution, and the horizontal axis is the number of days of storage. The concentration of the solution was dropped rapidly when the Aminosulfonic Acid Gemini Surfactant solution was stored for 1–2 days. On the third day, the concentration tended to be stable and unchanged basically. It can be seen that the concentration of the Aminosulfonic Acid Gemini Surfactant changes little when the temperature is 80°C. At 80°C, the resistance temperature of Aminosulfonic Acid Gemini Surfactant is the best [22].

4.2. The surface tension of Aminosulfonic Acid Gemini Surfactant solution

At 25°C, the surface tension (Table 1) of this Aminosulfonic Acid Gemini Surfactant in distilled water solution was measured by the ZL3000 automatic interfacial tension meter.

Table 2: Oil-water interfacial tension of the Aminosulfonic Acid Gemini Surfactant solutions.

SOLUTION CONCENTRATION (mmol/L)	INTERFACIAL TENSION (mN/m)
0.01	4.2
0.025	0.89
0.05	0.26
0.075	2.3×10^{-2}
0.1	1.2×10^{-3}
0.2	1.0×10^{-3}
0.5	9.8×10^{-4}
1	9.8×10^{-4}
2	9.7×10^{-4}
5	9.5×10^{-4}
10	9.4×10^{-4}

Figure 6: Sand filling pipe.

It is known from the table that the interfacial tension decreased with the increase of concentration. The downward trend of interfacial tension was slowed gradually. When the solution concentration reached 0.1 mmol/L, the surface tension was unchanged basically. It is known from the table that the newly synthesized Aminosulfonic Acid Gemini Surfactant has higher surface activity. The interfacial tension was reduced by adding this surfactant to the combination flooding system.

4.3. The oil-water interfacial tension of the Aminosulfonic Acid Gemini Surfactant solution

The entire experimental procedure is operated in accordance with the GB6541-86 "Petroleum Products Oil-to-Water Interfacial Tension Measurement Method". The oil-water interfacial tension (Table 2) of the Aminosulfonic Acid Gemini Surfactant solution and oil sands oil were measured by the interfacial tension measurement method.

It is known from the table that the minimum aqueous solution of the combination flooding system reaches an ultra-low level of 9.4×10^{-4} mN/m. They are all less than 10^{-3} mN/m at a certain concentration.

4.4. The combination flooding system of Aminosulfonic Acid Gemini Surfactant

The synthesized Aminosulfonic Acid Gemini Surfactant and polyacrylamide (HPAM) were used as the combination flooding system reagent [23–26]. When the concentration of HPAM is less than 1200 mg/L, the viscosity of the emulsion decreases with the increase of HPAM concentration. When the concentration of HPAM exceeds 1200 mg/L, the viscosity of the emulsion system increases with the increase of polymer concentration. Combined with the existing experimental conditions, HPAM of 1300 mg/L was selected. 1300 mg/L HPAM + 0.05% Aminosulfonic Acid Gemini Surfactant and 1300 mg/L HPAM + 0.1% Aminosulfonic Acid Gemini Surfactant were prepared respectively. Under the experimental temperature 80°C and flow rate 0.02–0.04 L/min, the composite flooding experiment (Figure 6) was carried out. Recorded the relevant data (Table 3) when the flow rate at the liquid outlet on the side of the sand-packing pipe was stabilized relatively.

Table 3: Oil sand combination flooding system scheme and test results.

TYPE	1300 mg/L (HPAM) + 0.05% AMINOSULFONIC ACID GEMINI SURFACTANT	1300 mg/L (HPAM) + 0.1% AMINOSULFONIC ACID GEMINI SURFACTANT
Temperature (℃)	80	80
Sand fill length (cm)	50	50
Sand core diameter (cm)	7.6	7.6
Cross-sectional area (cm^2)	45.36	45.36
Sand core weight (g)	960	960
Displacement period (h)	72	72
Liquid yield (ml)	94.6	102.23
Oil production (ml)	20.09	23.82
Water production (ml)	74.51	78.41
Oil recovery (%)	21.23	23.30

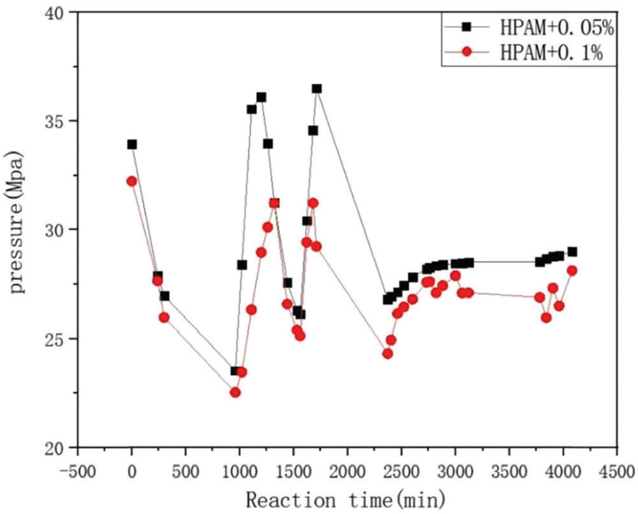

Figure 7: The changes of entrance pressures in complex drive experiment of oil sand.

4.4.1. The oil recovery rate changes of the combination flooding system

As can be seen from the above table. Carried out 1300 mg/L (HPAM) + 0.05% Aminosulfonic Acid Gemini Surfactant combination flooding system sand-packing pipe flooding experiment. The output liquid quality was 93.98 g. The volume of produced liquid was 94.6 mL. The volume of water produced was 74.51 mL. The oil production volume was 20.09 mL. The oil recovery rate was 21.23%. Carried out 1300 mg/L (HPAM) + 0.1% Aminosulfonic Acid Gemini Surfactant combination flooding system sand-packing pipe flooding experiment. The output liquid quality was 101.5 g. The volume of produced liquid was 102.23 mL. The volume of water produced was 78.41 mL. The oil production volume was 23.82 mL. The oil recovery rate was 23.30%.

4.4.2. The inlet and outlet pressure changes of the combination flooding system

The data statistics were performed about the inlet pressure (Figure 7) of the sand pack during the fluid output on one side of the sand pack was stabilized. The displacement time was the abscissa, and the inlet pressure was the ordinate.

It can be seen from the figure that the inlet pressure of 1300 mg/L (HPAM) + 0.1% Aminosulfonic Acid Gemini Surfactant solution is lower. The high concentration of Surfactants were showed excellent properties in lipophilicity and hydrophilicity [27].

The data statistics were performed about the outlet pressure (Figure 8) of the sand pack during the fluid output on one side of the sand pack was stabilized. The displacement time was the abscissa, and the outlet pressure was the ordinate.

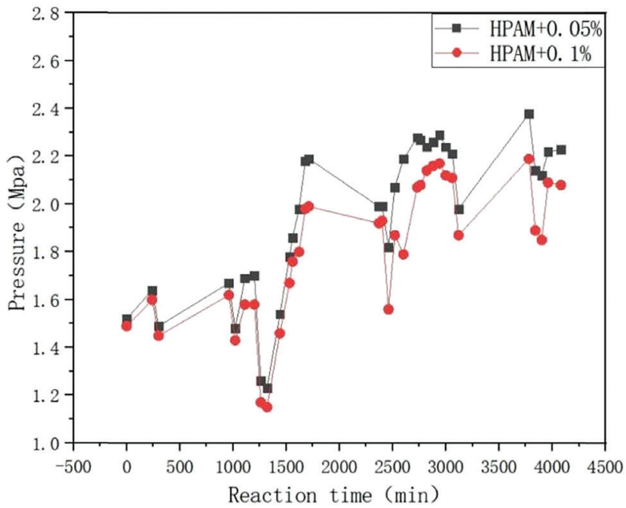

Figure 8: The changes of outlet pressures in complex drive experiment of oil sand.

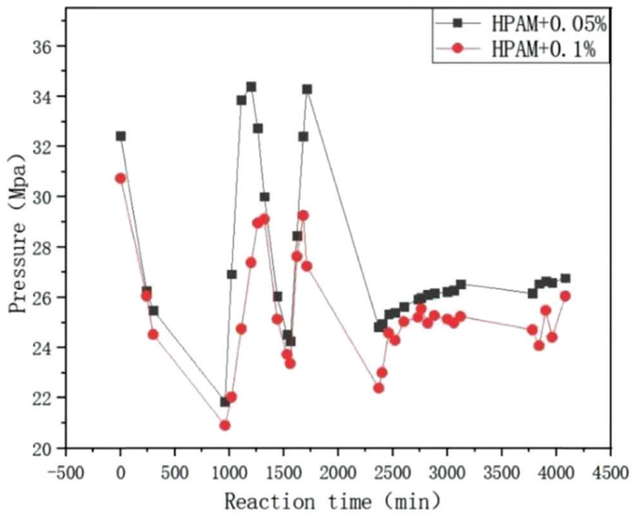

Figure 9: The changes of pressure difference in complex drive experiment of oil sand.

The results demonstrated that the outlet pressure of 1300 mg/L (HPAM) + 0.1% Aminosulfonic Acid Gemini Surfactant solution is lower. Not only the high-concentration of surfactants were showed excellent properties in lipophilicity and hydrophilicity, but also played a good role in improving permeability [28].

The data statistics were performed about the inlet and outlet pressure (Figure 9) of the sand pack during the fluid output on one side of the sand pack was stabilized. The displacement time was the abscissa, and the pressure difference between the inlet and outlet was the ordinate.

The pressure and permeability were fluctuated greatly during the displacement process. The pressure was risen when the oil sands just block the gap. It is obvious that the peak of pressure became larger with the increase of liquid. The smaller oil sands will be washed away from the gaps when a certain pressure is reached. At the same time, the permeability will rise briefly and the pressure will drop briefly [29–31].

4.4.3. Permeability changes of the combination flooding system

The data statistics were performed about the inlet and outlet pressure (Figure 10) of the sand pack during the experiment of the combination flooding system sand pack. The permeability was calculated according to the formula, the displacement time was the abscissa, and the composite flooding permeability was the ordinate.

The permeability of the combined flooding was improved when the Aminosulfonic Acid Gemini Surfactant solution was combined with polymer flooding. The penetration rate of the high concentration surfactant is better, which fully shows the promotion effect of the surfactant on the penetration [32–33].

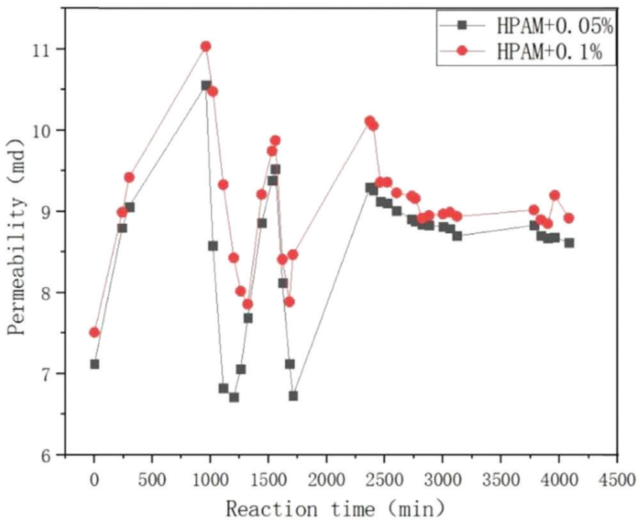

Figure 10: The changes of Permeability in complex drive experiment of oil sand.

At the beginning of the sand-packing pipe displacement, the permeability decreased. The interaction of the polymer and the sulfamic acid amphoteric Gemini surfactant quickly reduced the surface tension of the sample during the experiment, which increased the recovery rate. Due to the migration of oil sands, the liquid is displaced from one side during the displacement process, and the oil sands with smaller particles will also move to a certain extent. The pressure was risen when the oil sands just block the gap. Due to the continuous increase of liquid, the pressure will continue to rise. When a certain pressure is reached, the smaller oil sands will be washed away from those gaps, the permeability will rise briefly, and the pressure will be a brief drop.

5. CONCLUSION

A new AminoSulfonic Acid Gemini Surfactant has been synthesized, which is conducive to the research of material science. A new method is proposed for the research of surfactant oil displacement. This study mainly does the following research.

(1) A new type of the SNNDPE was synthesized with hexadecylamine and sodium 2-chloroethyl sulfonate. The SNNDPE has better reaction temperature at 80°C. The best reaction time of SNNDPE is 12 h. then the synthesized sample was verified by infrared spectroscopy. The structure of the SNNDPE was obtained.

(2) Performance testing: The Aminosulfonic Acid Gemini Surfactant solution has better resistance temperature at 80°C, so the 80°C was chosen as the experimental temperature of the combination flooding system. The surface tension of the sample test shows that its surface performance is more prominent than that of ordinary surfactants. Adding this surfactant to the compound flooding can reduce the interfacial tension very well. When the concentration of the solution reaches 0.1 mmol/L, the surface tension remains unchanged below 24.8 mN/m.

(3) Then we did the combination flooding system experiment. In the combination flooding system, Carried out 1300 mg/L (HPAM) + 0.05% Aminosulfonic Acid Gemini Surfactant combination flooding system sand-packing pipe flooding experiment. The output liquid quality was 93.98 g. The volume of produced liquid was 94.6 mL. The volume of water produced was 74.51 mL. The oil production volume was 20.09 mL. The oil recovery rate is 21.23%. Carried out 1300 mg/L (HPAM) + 0.1% Aminosulfonic Acid Gemini Surfactant combination flooding system sand-packing pipe flooding experiment. The output liquid quality was 101.5 g. The volume of produced liquid was 102.23 mL. The volume of water produced was 78.41 mL. The oil production volume was 23.82 mL. The oil recovery rate is 23.30%.

(4) The combination flooding system phenomenon shows that the surface activity of the polymer flooding is greatly increased and the interfacial tension is reduced. The ultra-low interfacial tension was reduced by the combination of Aminosulfonic Acid Gemini Surfactant and polymer. The combination flooding system produces more fluids that Compared with water flooding and polymer flooding. In laboratory experiments, the Aminosulfonic Acid Gemini Surfactant solution has a obvious effect on enhancing oil recovery.

In summary, the combination flooding system has the best Oil recovery rate effect. The author believes that this kind of alkali-free oil displacement system not only avoids the problems of bottom-hole scaling and pump stuck in production wells caused by alkali in the oil displacement process, but also achieve ultra-low interfacial tension and high viscosity. The above results can be considered as a guidance in the design and studies of the combination flooding system for a selective. As for the above the combination flooding system, there are few experiments at present. The experimental data are not sufficient. I hope later scholars can do more experiments in this field.

6. ACKNOWLEDGEMENTS

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