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Evaluation on the Basis and Application Performance of a Modified Surface Active Oil-displacing Agent

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Abstract: *The oil development has entered the water flooding stage in Changqing Oilfield. Due to the low reservoir permeability and the high oil viscosity, the water-flooding does not achieved the desired effect. A large amount of oil is retained in the reservoir due to the high water surface tension and oil-water interfacial tension. Currently, the performance of an oil-displacing agent is suitable for Changqing oilfield to mitigate this situation, but with high application cost due to its synthesis in an organic solution. In this study the oil-displacing agent is modified, and an amphiphilic oil-displacing agent is synthesized through aqueous polymerization. With addition of the oil-displacing agent, the surface/interface activity of water and oil changed significantly. The surface tension of water and oil reduces from 69mN/m to 25mN/m and from 21mN/m to 7.8mN/m respectively in the 1500mg/L oil-displacing agent at 60℃, and the oil-water interfacial tension reaches the ultra-low level. In addition, the oil-displacing agent has the good oil emulsification ability, and the emulsion shows stable performance. The core flooding experiment shows that the 2000mg/L oil-displacing agent can increase the oil recovery by 20% compared with the water flooding, and this meets the production needs in the oilfield. Since the oil-displacing agent contains a large number of chelating functional groups, so it can also chelate the heavy metal ions in the reservoir to prevent precipitation with incompatible anions in the injected water.*

Keywords**:** *oil-displacing agent, performance evaluation, core flooding experiment*

1. Introduction

With the growing demand for energy resources and the continuous reduction in production of conventional resources, more attention has been paid to the exploitation of unconventional resources, among which heavy oil and residual oil are most promising [1]. Heavy oil and residual oil are difficult to effectively recover by water-flooding [2]. China's water-flooding developed oilfields represent the highest proportion in the world and most of oilfields have entered the high water cut stage. Most oilfields in China, especially the Changqing Oilfield, have complex geological conditions, great differences in oil properties and serious reservoir heterogeneity. Evaluation shows the viscosity of oil is 423mPa·s at surface and 53mPa·s in the 1.5mD reservoir at 5-6ºC at the depth of 1500m-2000m in Changqing Oilfield, where the formation water has the salinity of 1026.05mmol/L and contains carbonate, sulfate, calcium ions, etc. Currently, the Changqing Oilfield has entered the water flooding development stage. However, the water cannot flow into the reservoir pores due to the high surface tension. After water flooding, 60%-70% of oil is still retained as the residual oil [3].

The common oil displacement technologies to improve the oil recovery include CO_2 flooding, alkaline flooding and oil-displacing agent (ODA) flooding. For the heavy oil with the high colloid content, CO_2 will destroy the colloid structure and cause asphaltenes coagulate, which blocks the reservoir pores. The mechanism of alkaline flooding requires alkaline solution to react with the acidic macromolecules in the oil to result in surfactants. However, this method does not meet the conditions at Changqing Oilfield, since the heavy oil has the complex compositions, and its properties are different at different depths. Among those techniques, the ODA is one of the most feasible methods to improve the oil recovery in Changqing Oilfield by adding amphiphilic emulsifying active agents into heavy oil, due

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to its good compatibility and excellent oil displacement efficiency, which can realize with the low concentration and under high salinity conditions [4-7].

Currently, ODAs are widely applied in oil fields. Changxin Shi (2017) [8] used erucami-dopropyl dimethyl amine, ethylenediaminetetraacetic acid disodium salt and hydrolyzed polyacrylamide to obtain the surfactant of erucamidopropyl dimethyl amine oxide. The oil-water dynamic interfacial tension was evaluated. The surfactant has the best effect on reducing the surface tension with the concentration of 0.1%. After 20 min, the oil-water dynamic interfacial tension reaches 0.001mN/m, which is ultra-low, but the interfacial tension increases gradually over time. Babu et al (2016) [9] synthesized a polymer surfactant with castor oil and acrylamide, and the core flooding experiment showed that the oil recovery reaches 26.5%, 27.8% and 29.1% respectively when the concentration is 0.5%, 0.6% and 0.7%. Gui et al (2008) [10] synthesized octylphenol polyoxyethylene ether maleate monoester (OPMA) and then made P (AM-co-OPMA) polymer through copolymerization of acrylamide with potassium persulfate sodium bisulfite as initiator. The polymer has high thickening ability, robust surface activity and interfacial activity and can be used as a ODA in oilfield. Juan Li (2019) [11] synthesized a new ODA AAGAS at low temperature. The evaluation shows that the ODA has the good surface and interfacial activity, and the oil-water interfacial tension is ultra-low in the 1000mg/L ODA, which effectively reduces the water surface tension and the oil viscosity. Through literature research in this field, the performance of existing ODA is quite different and the effect is inconsistent for different properties of injected water and crude oil. Therefore, a new ODA is developed for the reservoir and crude oil properties of Changqing Oilfield, and the production process is controlled to achieve the goal of reducing production costs.

AADAS ODA mentioned in the literature which developed by Juan Li is applicable in Changqing Oilfield. AAGAS ODA is made with copolymerizing acrylamide, 2-acrylamido-2-methylpropane sulfonic acid and glycidyl methacrylate to obtain a reactive copolymer, and then the acrylamide-sodium styrene sulfonate binary copolymer is grafted onto the reactive copolymer through the ring-opening reaction between the epoxy group and the amino group. At the temperature of 6^{o}C and with the 2000 mg/L AAGAS ODA, the surface tension of water reduces from 65 mN/m to 32 mN/m and the oil-water interfacial tension reduces to below 0.001mN/m, which is ultra-low. The monomers selected for synthesis are all common reactive monomers, but the synthesis of the AAGAS copolymer requires dimethyl sulfoxide as the solvent, which leads to the high cost and the complicated operation [12]. Moreover, the AAGAS ODA is composed of amino groups and epoxy groups, and during synthesis, the ring-opening reaction of the epoxy group may cause too long molecular chain and the higher viscosity. The high viscosity of the ODA is not conducive to its diffusion in the reservoir pores [13]. In this study, to prevent the high viscosity, the acrylamide used to synthesize the copolymer is replaced with maleic anhydride and dimethyl maleate, and aqueous solution polymerization is used to reduce the synthetic cost [14] and a new ODA named DMAG-AS is synthesized. The surface/interfacial activity, crude oil emulsification, viscosity reduction, and core flooding experiments of the ODA are performed on it, and its performance is compared with AAGAS ODA to determine whether it meets the needs of the application.

2.Materials and methods

2.1. Evaluation of the basic properties of DMAG-AS ODA

2.1.1. Evaluation of the surface activity of DMAG-AS ODA

Surface activity is the main factor affecting the ODA performance. Excellent surface activity reduces the surface tension of the aqueous solution, makes easier flow in the reservoir pores and enhances the sweep efficiency. As the surface tension of the oil decreases, the oil becomes easier to disperse and can carried by the ODA solution effectively, which improves the oil displacement efficiency of the ODA on crude oil [15]. To evaluate the performance of ODA, the effects of DMAG-AS ODA on the surface tension of water and oil are evaluated first.

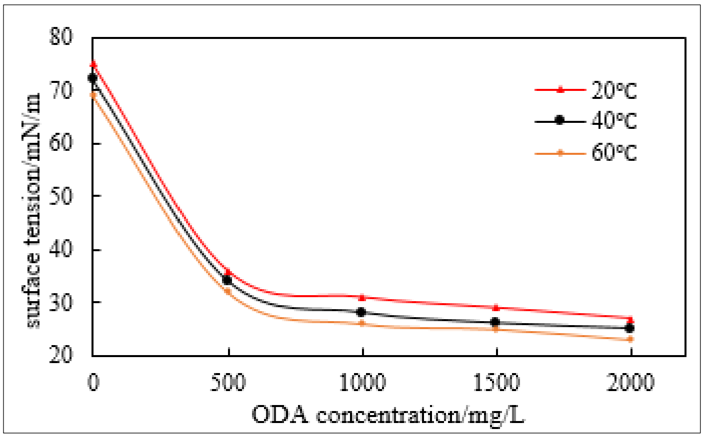


Figure 1. Effect of different concentrations of DMAG-AS polymer flooding agent on water surface tension at different temperatures

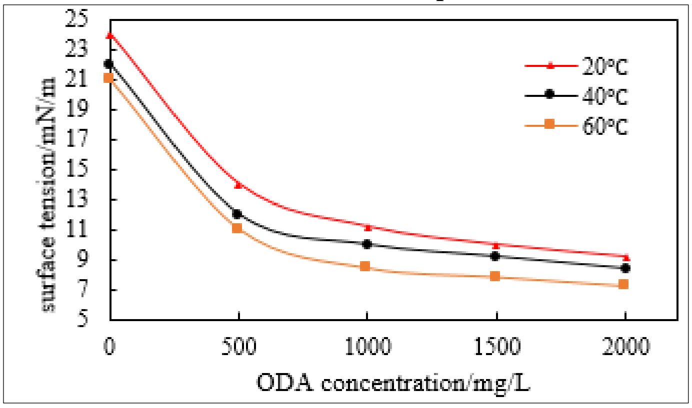
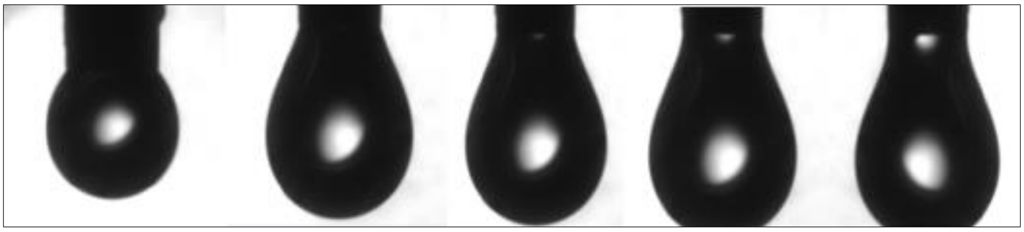


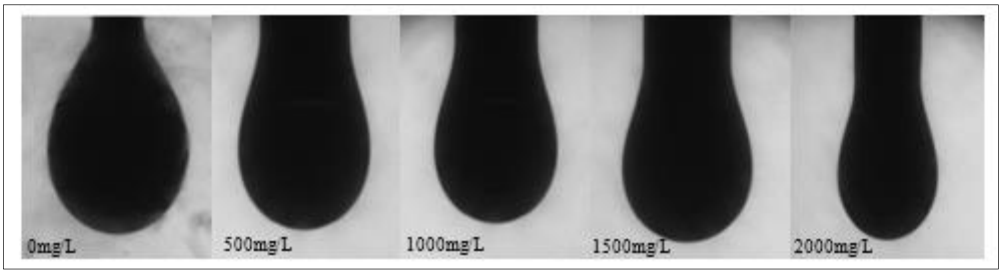
Figure 2. Effect of concentrations of DMAG-AS polymer ODA on oil surface tension at different temperatures

The surface tension of water and oil at different temperatures and different concentrations of ODA are evaluated. As shown in Figure 1 and Figure 2, the DMAG-AS ODA has a good performance to reduce the surface tension of oil and water, and the effect of ODA to reduce surface tension improves as temperature increases. When the temperature is 20°C, the surface tension of water and oil reduce from 75mN/m and 24mN/m to 34mN/m and 14mN/m in the 500mg/L ODA solution, and the surface tension decrease to 25mN/m and 9.2mN/m when concentration of the solution goes up to the 2000 mg/L. When the temperature increases to 60°C, the surface tension of water and oil reduce to 23mN/m and 7.2mN/m in the 2000mg/L ODA solution. In the same situation, AAGAS ODA can only reduce the surface tension of water to 32 mN/m, so the synthetic new ODA has better surface activity.



(a) (b) (c) (d) (e)

Figure 3. The morphology of water droplets in the air varies with different concentration of DMAG-AS ODA**.** (a, b, c, d, e are the droplet morphology of DMAG-AS ODA aqueous solution in the air at 40 °Cwith the concentrations of 0 mg/L, 500 mg/L, 1000 mg/L, 1500 mg/L and 2000 mg/L respectively)



(a) (b) (c) (d) (e)

Figure 4. The morphology of oil droplets in the air varies with different concentration of DMAG-AS ODA. (a, b, c, d, e are the morphology of oil droplets with concentrations of DMAG-AS polymer ODA of 0 mg/L, 500 mg/L, 1000 mg/L, 1500 mg/L and 2000 mg/L respectively at 40℃)

Figure 3 and Figure 4 shows the morphology of water droplets and oil droplets containing different concentrations of DMAG-AS ODA in the air. When no oil-displacing agent is added, the water droplets and oil droplets are spherical due to the effect of surface tension. With the increasing ODA concentration, the shapes of water droplets and oil droplets show the same trend of change that the shapes change from spherical to ellipsoidal or even strip. This phenomenon also shows that the obtained DMAG-AS ODA can significantly reduce the surface tension of oil and water, and has good surface activity.

2.2. Interfacial activity analysis of DMAG-AS ODA

The oil-water interfacial tension is an important parameter affecting the exploitation of heavy oil and residual oil. Under the low interfacial tension, the oil is more easily to disperse in the solution to form droplets, which causes transition from oil high friction force to low friction force between oil phase and aqueous phase [16]. The oil is more easily carried by water and has the better mobility in the reservoir, which improves the oil recovery factor [17]. The interfacial activity of the ODA with different concentrations at various temperatures and the morphology of oil droplets in different concentrations of the DMAG-AS ODA solutions are evaluated, as shown in Figure 5 and Figure 6.

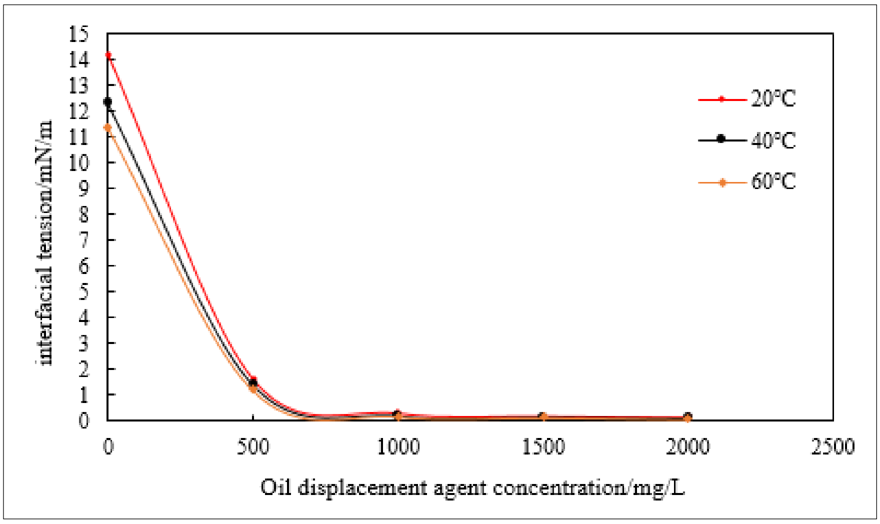


Figure 5. Oil-water interfacial tension of DMAG-AS polymer ODA with different concentrations at different temperatures

We notice that the DMAG-AS ODA effectively reduces the oil-water interfacial tension. When the temperature increases to 60°C, the oil-water interfacial tension reduces to 0.0014 mN/m in the 1000 mg/L ODA solution and to 0.0008 mN/m in the 1500 mg/L ODA solution, realizing ultra-low interfacial tension. The morphology of oil droplets change in different concentrations of ODA solutions, and the

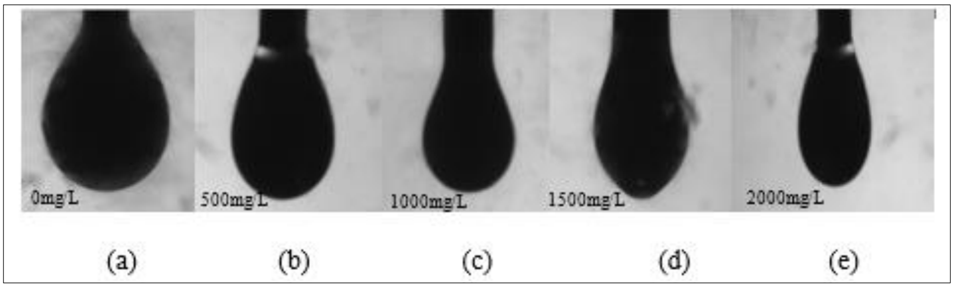


Figure 6. The morphology of oil droplets in DMAG-AS ODA aqueous solution with different concentrations**.** (a, b, c, d, e are the morphology of oil droplets in DMAG-AS ODA aqueous solution with the concentrations of 0 mg/L, 500 mg/L, 1000 mg/L, 1500 mg/L and 2000 mg/L respectively at 40°C)

degree of change increases with the increase of ODA concentration. The DMAG-AS ODA can significantly reduce the oil-water interfacial tension through evaluation experiments. Similarly, when the AAGAS ODA concentration is higher than 1000 mg/L, the oil-water interfacial tension drops below 0.001 mN/m. It is found that the interfacial activity of the ODA in this study is close to that of AAGAS ODA.

In summary, the DMAG-AS ODA is obtained by using maleic anhydride and dimethyl maleate to replace the acrylamide in the synthesis of AAGAS ODA. Because of maleic anhydride and dimethyl maleate, the product contains more hydrophilic groups and is more easily to adsorb on the water surface. Compared with the AAGAS ODA, the DMAG-AS ODA has better oil-water surface activity and similar oil-water interface activity under the premise of reducing production costs and processes.

3. Evaluation of effects on oil displacement ability of DMAG-AS ODA

The oil displacement ability of the ODA is the result of the combined effect of many factors, such as the diffusion capacity of injected water, the emulsification and viscosity reduction ability of the ODA to crude oil, etc. [18]. Each factor is the combination of different properties of ODA. Through the evaluation, the DMAG-AS ODA has good surface/interface activity, so the ability of the ODA to affect the oil-displacing effect is evaluated.

3.1. Core diffusion experiment of DMAG-AS ODA solution

As the DMAG-AS ODA can significantly reduce the surface tension of water, low surface tension makes water easier to flow into the pores of low permeability reservoirs. To confirm this theory, a core diffusion experiment is carried out. Select a core with a permeability of 1.5mD and take a core slice with a thickness of 0.3mm, and place it in ODA aqueous solution with a concentration of 1000mg/L to observe the degree of dispersion of the ODA solution in the rock slices within 4 h.

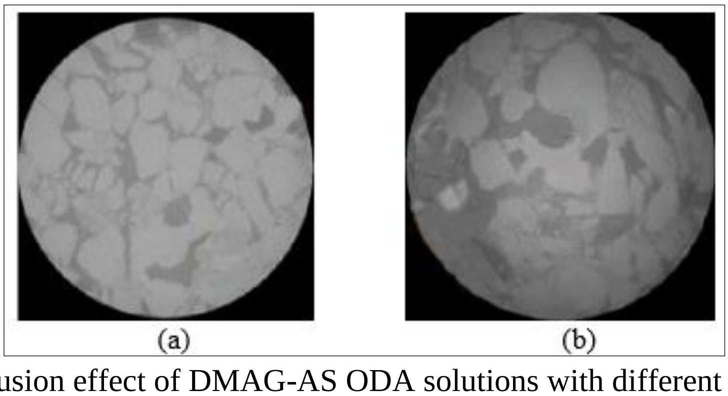


Figure 7. Diffusion effect of DMAG-AS ODA solutions with different concentrations in rock slices (a is the diffusion effect of a solution that does not contain oil-displacing agent for 4 h, b is the diffusion effect of a solution of oil displacing agent with a concentration of 1000mg/L in 4 h)

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It can be seen from the evaluation result of Figure 7, due to the addition of the ODA, the diffusion capacity of water in the rock slices has been significantly improved due to the reduction of the surface tension of the water. Therefore, in the process of crude oil extraction, adding the DMAG-AS ODA can significantly increase the sweep coefficient of the injected water in the reservoir and increase the recovery factor.

3.2. Evaluation of the oil emulsification ability of DMAG - AS ODA

The DMAG-AS ODA has good interfacial activity so it can be adsorbed on the oil-water interface to form an emulsion, which effectively reduces the oil apparent viscosity and the oil mobility, which lead the ability of water to carry oil is enhanced [19]. To evaluate the viscosity-reducing effect of the obtained ODA on crude oil in Changqing Oilfield, the viscosity of oil in Changqing Oilfield is measured firstly. Then, the oil emulsification experiment is operated at different temperatures to observe the trend of the apparent viscosity as the ODA concentration increases. Finally, the morphology of the formed emulsion droplet is observed, and the stability of the emulsion is evaluated according to the droplet morphology [20].

Table 1. Oil viscosity in Changqing Oilfield

Temperature/°C	Oil viscosity/mPa·s
20	435.2
40	263.3
60	152.8
80	52.3

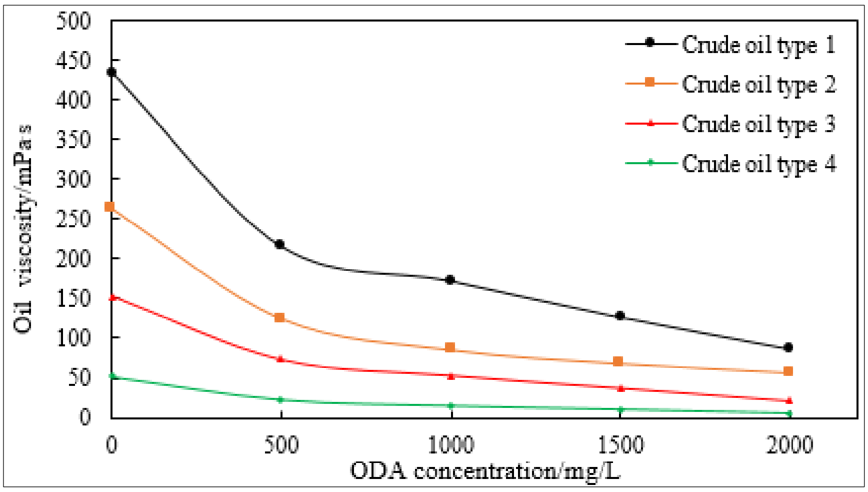


Figure 8. Effect of polymer ODA concentration on the oil viscosity

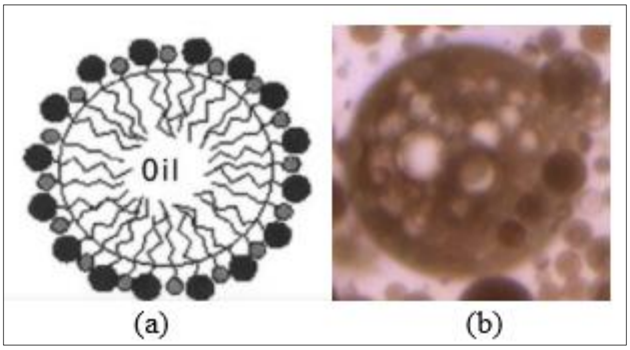


Figure 9. Schematic diagram of oil-in-water emulsion and emulsion droplet morphology (a is the Schematic diagram of oil-in-water emulsion, b is the emulsion droplet morphology)

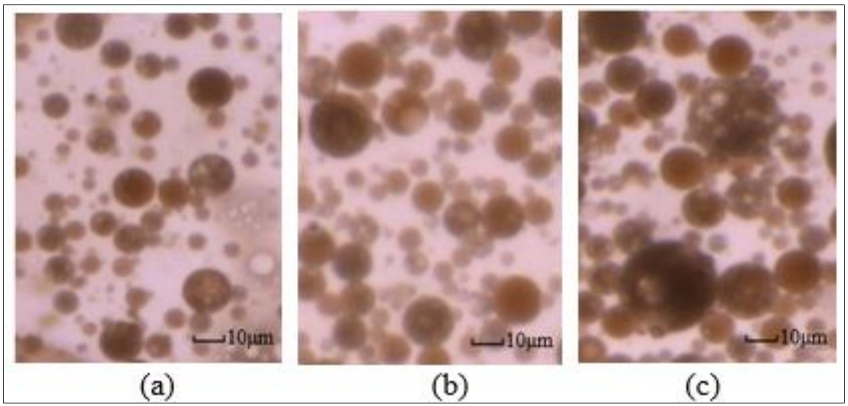


Figure 10. The form of crude oil emulsion at different times
（a, b, c are the morphology of crude oil emulsion at 1 h, 6 h and 12 h, respectively）

As shown in Figure 8, the 1500mg/L ODA significantly reduces the oil viscosity in Changqing Oilfield. At the reservoir temperature of 60ºC, due to the emulsification effect of ODA on crude oil, the oil viscosity reduces from 152.8 mPa·s to 38.2 mPa·s, which is in the range of thin oil [21]. By observing the structure of the emulsion droplets in Figure 9, it is obtained that DMAG-AS ODA solution and crude oil form an oil-in-water emulsion, and oil-in-water emulsion can effectively reduce the viscosity of crude oil. It can be seen from the morphology of emulsion droplets at different times in Figure 10, most of the droplets with a standing time of 1 h have a particle size of less than 10 μm. The particle size of the emulsion increases with the standing time, when the standing time increases to 6 h, the number of droplets larger than 10 μm in the emulsion increases significantly. When the standing time increases to 12 h, large-size emulsion droplets appeared in the emulsion, it shows that some emulsion droplets have coalesced. Although the particle size uniformity of the obtained emulsion drops continuously decreases with the increase of the standing time, its stability can meet the needs of oilfield production.

3.3. Analysis of DMAG-AS ODA performance in core flooding

Based on the conclusions of the above evaluation experiments, the cores of Changqing Oilfield are selected to use for core flooding experiments with different concentrations of DMAG-AS ODA. The oil displacement ability of DMAG-AS ODA is evaluated by the recovery factor of the crude oil in the core, and is compared with the recovery factor of water flooding and AAGAS ODA. At first, the 1 cm long core and 7 cm core long are saturated with the 152mPa·s and 53mPa·s oil at 60℃ respectively to evaluate the displacement effect of different ODAs. Then, the deuterium oxide is used to operate the flooding experiment and evaluate the recovery factor under different concentrations of different ODAs, and compare the results with the recovery factor in the conventional water-flooding. Finally, the law of core flooding is observed by nuclear magnetic imaging.

Table 2. Comparison of oil recovery between DMAG-AS ODA flooding and water flooding

No.	Polymer concentration(mg/L)	Recovery rate of water flooding (%)	Recovery rate of polymer flooding (%)
1	500	26.2	34.7
2	1000	26.2	37.5
3	1500	26.2	41.3
4	2000	26.2	45.4

Table 3. Comparison of recovery efficiency between DMAG-AS and AAGAS ODAs

Agent concentration	Enhanced oil recovery factor (%)	
	DMAG-AS	AAGAS
1000mg/L	11.3	9.3
1500mg/L	15.1	11.4
2000mg/L	19.2	14.5

As stated in Table 2 and Table 3, the recovery factor in conventional water flooding is only 26.2%, which is caused by the low permeability and the clay swelling in cores. Compared to the water flooding, the 2000mg/L DMAG-AS ODA leads to the recovery factor up to 45.4%, which increases by nearly 20%. Under the same conditions, DMAG-AS ODA has a higher recovery factor than that of AAGAS ODAs, and enhances the oil recovery.

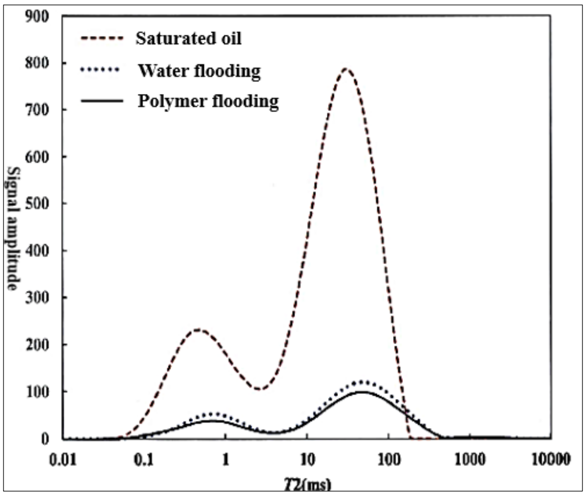
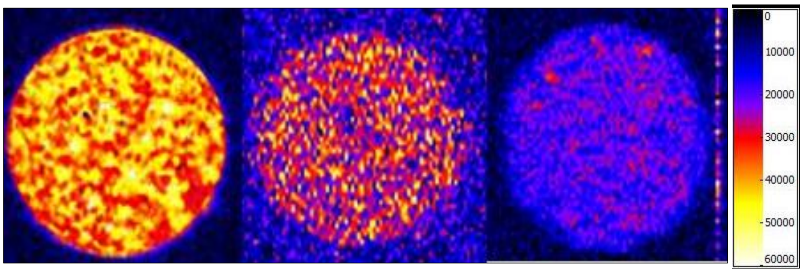
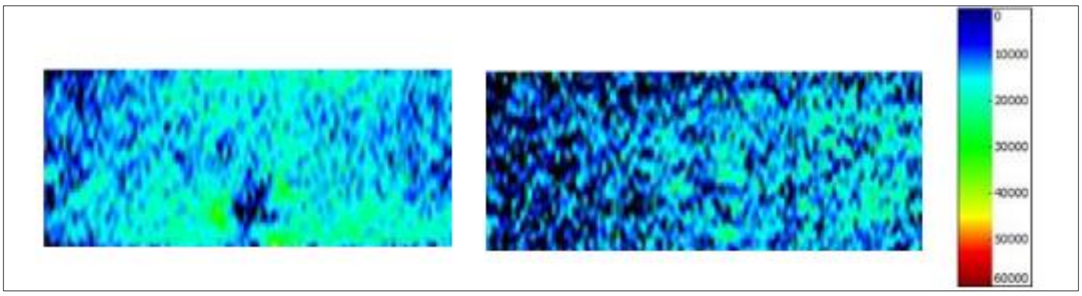


Figure 11. T2 spectrum of polymer flooding experiment



(a) (b) (c)

Figure 12. The nuclear magnetic resonance imaging of 1 cm long core entrance with different ODAs (a is the nuclear magnetic resonance imaging of 1 cm long saturated oil core entrance, b is the nuclear magnetic resonance imaging of the cores-entrance of water-driven, c is the nuclear magnetic resonance imaging of the cores-entrance of DMAG-AS ODA driven)



(a) (b)

Figure 13. The nuclear magnetic resonance imaging of 7 cm long core with different ODAs (a is the nuclear magnetic resonance imaging of the core of water-driven, b is the nuclear magnetic resonance imaging of the core of DMAG-AS ODA driven)

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The relationship between T_2 relaxation time and pore size in nuclear magnetic resonance was calculated according to Equation (1)

$$\frac{1}{T_2} = \rho_2 \frac{S}{V} + \frac{\gamma^2 G^2 D^2}{3} \qquad (1)$$

where *D* is the diffusion coefficient, *G* is the magnetic field gradient, γ is the gyromagnetic ratio, S/V is the pore specific surface, and ρ_2 is the transverse surface relaxation strength [22].

It can be seen that T_2 relaxation time is proportional to the pore radius. The T_2 spectrum of nuclear magnetic resonance after water flooding and polymer flooding are shown in Figure 11 and the results shows the T_2 range of the polymer flooding is larger than that of the saturated oil core. The deuterium oxide is used in the experiments, and the pore structure of the core does not change significantly during the process of the experiments, which is due to emulsion [23]. As shown in the nuclear magnetic resonance imaging of core entrance in Figure 12 and Figure 13, water flooding can only displace a small part of the oil in the core and the displacement capacity is limited. Compared to the water flooding, the polymer flooding displaces oil in core more effectively. It is concluded that DMAG-AS ODA has an enhanced effect of improving oil recovery.

It can be concluded that DMAG-AS ODA can reduce the oil viscosity through emulsification and improve the oil displacement efficiency in water flooding due to its robust surface/interface activity. Therefore, the DMAG-AS ODA can significantly improve the recovery efficiency of residual oil and heavy oil in the reservoir pores.

3.4 Evaluation of the chelating and anti-scale ability of DMAG-AS ODA

The 2-acrylamide-2-methylpropanesulfonic acid and maleic anhydride are used as synthetic monomers, and the molecular structure of the product is composed of chelating functional groups such as –OH and -COOH. The water used in water flooding in Changqing Oilfield contains sulfate and carbonate, and heavy metal ions such as Ca^{2+}, Ba^{2+} and Sr^{2+} are present in the reservoirs [24]. Thus, precipitation always occurs when the water is injected into the reservoir, causing blockage to the reservoir pores. The chelating functional group in the DMAG-AS ODA can chelate the heavy metal ions in the formation to prevent scaling, and the DMAG-AS ODA is used to evaluate the scale inhibition rate at different temperatures and ODA concentrations with the scaling ionic concentration of 600 mg/L. Furthermore, the effect of polymer on the crystal morphology is observed by adding ODA in $BaSO_4$ crystal growth stage.

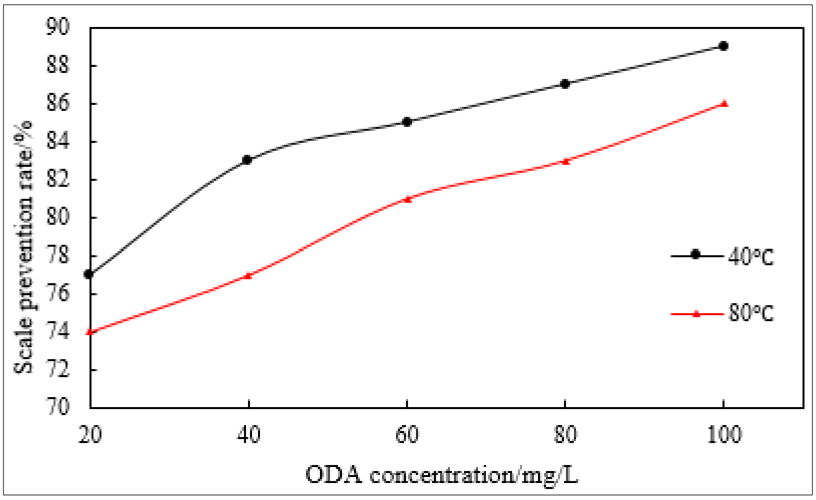

Figure 14. Scale inhibition rate of ODAs

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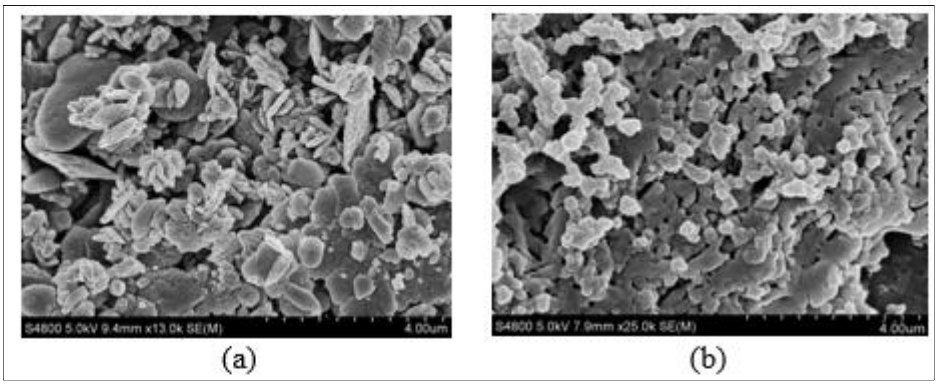

Figure 15. The effect of DMAG-AS on the structure of barium sulfate scale (a is the microscopic morphology of barium sulfate without ODA, b is the microscopic morphology of barium sulfate after adding ODA)

Based on the Figure 14, the ODA has the good anti-scaling ability, which enhances with the increase of its concentration. At the temperature of 60ºC, the 20 mg/L ODA has the scale inhibition rate of 77%, and the 100 mg/L ODA has the scale inhibition rate of 89%. The ODA has the good temperature resistance due to its sulfonic groups. At the experimental temperature of 80ºC and the ODA concentration of 100 mg/L, the scale inhibition rate reduces but still maintains above 85%. As shown in Figure 15, without adding ODA, barium sulfate crystals accumulate to form rosette shaped or bifurcated crystal blocks, and the arrangement structure of it is tight and irregular. After adding ODA, the normal growth of crystal is affected by functional groups with chelating ability and the barium sulfate crystal turns to be spherical, and the structure is regular, which is more conducive to the mechanical cleaning or chemical scale dissolving in the later stage. To sum up, DMAG-AS ODA prevents the scaling due to the in-compatibility between the injected water and the formation heavy metal ions.

4. Conclusions

In this study, we successfully synthesized DMAG-AS ODA by modifying an existing oil-displacing agent. The new ODA is obtained by aqueous solution polymerization technology, which has lower cost and simpler production process. We demonstrated that DMAG-AS ODA has the good performance to reduce the surface tension and interfacial tension. At the temperature of 60ºC and with the 1000 mg/L ODA, the surface tension of water and oil reduces from 69 mN/m to 26 mN/m and from 21 mN/m to 8.4 mN/m respectively. In the 1500 mg/L ODA, the oil-water interfacial tension reduces to 0.0008mN/m, which is ultra-low. In addition, the 1500mg/L ODA significantly reduces the oil viscosity in Changqing Oilfield. The ODA and oil can form the stable emulsion with regular droplet structure, and the emulsions remain stable within 12 h. It shows that the polymer ODA has the good oil emulsification and viscosity reduction ability. The oil displacement experiment is operated with the 53mPa·s oil in cores of Changqing Oilfield. The recovery factor in water flooding is 26.2% and increases to 45.4% in the 2000mg/L ODA. The results are also confirmed in NMR imaging that emulsion is formed during the displacement process. Furthermore, the ODA is composed of hydroxyl and carboxyl groups with chelating ability and has the ability to prevent scale in pipeline and formation. At the reservoir temperature of 60ºC, the 100 mg/L ODA has the scale inhibition rate of 89%. The polymer causes the loose structure of scale particles, which are easily treated.

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