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Original article

Emulsification Stability of An Amphiphilic Polymer for Chemical Flooding

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ABSTRACT

Background: Emulsification plays a pivotal role in the process of enhanced oil recovery, especially in chemical flooding. Emulsification has emerged as one of the key mechanisms facilitating oil recovery in polymer flooding.

Aim: This study aimed to solve the problem of emulsification and stability of amphiphilic polymers in the process of oil displacement.

Materials and methods: The emulsion was prepared by stirring emulsification method in the lab, and the dynamic stability of the emulsion was determined by stabilizer, and the size and distribution of droplets were determined by laser particle sizing instrument.

Results: The experimental results show that, with the increasing mass concentration of amphiphilic polymer, the apparent viscosity of the solution is significantly increased. The emulsification ability and the stability of the emulsion are also enhanced. In addition, the microstructure of the emulsion shows that the amphiphilic polymer with higher concentration helps to reduce the particle size of the emulsified oil droplets and impels the more uniform distribution. Furthermore, the amphiphilic polymer system was conductive to improving the oil-water emulsification ability and prolonging the stability of the emulsion, especially in high-salinity and high-temperature environments.

Conclusion: The results of the study are of guiding significance for the emulsification of amphiphilic polymers for oil recovery.

Keywords: *amphiphilic polymer; emulsification; stability; rheology.*

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Оригинальное исследование

Устойчивость амфифильного полимера к эмульгированию при химическом заводнении

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АННОТАЦИЯ

Обоснование. Эмульгирование играет ключевую роль в процессе повышения нефтеотдачи, особенно при химическом заводнении. Эмульгирование стало одним из ключевых механизмов, способствующих извлечению нефти при полимерном заводнении.

Цель. Данное исследование было направлено на решение проблемы эмульгирования и стабильности амфифильных полимеров в процессе вытеснения нефти.

Материалы и методы. Эмульсию готовили методом эмульгирования при перемешивании в лаборатории, динамическую стабильность эмульсии определяли с помощью стабилизатора, а размер и распределение капель определяли с помощью лазерного прибора для определения размера частиц.

Результаты. Результаты экспериментов показывают, что с увеличением массовой концентрации амфифильного полимера кажущаяся вязкость раствора значительно увеличивается. Также повышается способность к эмульгированию и стабильность эмульсии. Кроме того, микроструктура эмульсии показывает, что амфифильный полимер с более высокой концентрацией помогает уменьшить размер частиц эмульгированных капель нефти и способствует их более равномерному распределению. Кроме того, амфифильная полимерная система способствовала улучшению способности эмульгирования нефти с водой и продлению стабильности эмульсии, особенно в условиях высоких солености и температуры.

Заключение. Результаты исследования имеют решающее значение для эмульгирования амфифильных полимеров при добыче нефти.

Ключевые слова: *амфифильный полимер, эмульгирование, стабильность, реология.*

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Түпнұсқа зерттеу

Химиялық суландыру кезінде амфифилді полимердің эмулгацияауға төзімділігі

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АННОТАЦИЯ
Негіздеу. Эмулгациялау мұнай өндіруді арттыру процесінде, әсіресе химиялық суландыру кезінде шешуші рөл атқарады. Эмулгациялау полимерлі су тасқыны кезінде мұнай алуға ықпал ететін негізгі механизмдердің біріне айналды.
Мақсаты. Бұл зерттеу мұнайды ығыстыру процесінде амфифилді полимерлердің эмулгациялауы мен тұрақтылығы мәселесін шешуге бағытталған.
Материалдар мен әдістер. Эмульсия зертханада араластырылған кезде эмулгациялау әдісімен дайындалды, эмульсияның динамикалық тұрақтылығы тұрақтандырғышпен анықталды, ал тамшылардың мөлшері мен таралуы бөлшектердің мөлшерін анықтау үшін лазерлік құралмен анықталды.
Нәтижелері. Тәжірибе нәтижелері амфифилді полимердің массалық концентрациясының жоғарылауымен ерітіндінің айқын тұтқырлығы айтарлықтай артады деп болжайды. Эмулгациялау қабілеті мен эмульсияның тұрақтылығы да артады. Сонымен қатар, эмульсияның микроқұрылымы жоғары концентрациясы бар амфифилді полимер эмулгацияланған мұнай тамшыларының бөлшектерінің мөлшерін азайтуға көмектесетінін және олардың біркелкі таралуына ықпал ететінін көрсетеді. Сонымен қатар, амфифилді полимер жүйесі мұнайдың сумен эмульгациялау қабілетін жақсартуға және эмульсияның тұрақтылығын ұзартуға ықпал етті, әсіресе жоғары тұздылық пен температура жағдайында.
Корытынды. Зерттеу нәтижелері мұнай өндіруде амфифилді полимерлерді эмулгациялау үшін өте маңызды.

Негізгі сөздер: *амфифилді полимер, эмулгациялау, тұрақтылық, реология.*

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Introduction

As one of the most important global energy sources, oil supports the economic development and industrial production of modern society [1]. However, with the gradual decrease of global oil reserves, the recovery rate of conventional oil recovery techniques is gradually decreasing, and the extraction of residual oil becomes more difficult. To improve the recovery rate, chemical flooding technology, as a key method in tertiary oil recovery (EOR), significantly improves the recovery rate by reducing the interfacial tension between oil and water and changing the fluidity ratio [2].

In chemical oil flooding, polymer oil flooding technology increases the viscosity of the water phase by introducing polymers to increase the oil-water flow ratio and improve the reservoir wave coefficient, thus improving the recovery of crude oil [3]. However, traditional polymers such as partially hydrolyzed polyacrylamide (HPAM) have poor properties in high-temperature and high-salinity environments, thus resulting in their limited application in complex reservoirs. In recent years, researchers have turned their attention to amphiphilic polymers with both hydrophilic and hydrophobic properties [4]. Amphiphilic polymers not only have better temperature and salt resistance but also can form a three-dimensional network structure through the self-assembly of their hydrophobic groups, which significantly improve the rheological properties and stability of the solution [5]. Seright et al. pointed out that the introduction of amphiphilic polymers effectively solves the viscosity reduction problem of traditional polymers in highly mineralized reservoirs, and significantly improves their shear and salt resistance [6]. The amphiphilic polymers have better application prospects in EOR technology in high-salinity reservoirs due to their excellent properties.

In EOR technology, the complex system of amphiphilic polymers and surfactants can significantly improve the emulsification ability [7]. The stability of emulsion directly affects the oil flooding efficiency, especially in high-temperature and high-salinity reservoirs [8]. Levitt et al. pointed out that, by introducing amphiphilic polymers into the solution, the interfacial tension between oil and water can be effectively reduced and a stable emulsion can be formed, which improves the dispersibility and mobility of oil and water [9]. In addition, Negin et al. showed that amphiphilic polymers effectively prevented oil droplet agglomeration through the formation of hydrophobic associative structures by self-assembly, thus dramatically improving the stability of emulsions [8]. Kang Wanli (1997) pointed out that salinity and temperature-resistant amphiphilic polymers had an important influence on emulsion stability in high mineralization environments [2].

In high-salinity environments, the introduction of electrolytes compresses the double layer, leading to accelerated oil droplet aggregation and decreased emulsion stability [10]. However, Negin et al.'s experimental results showed that the special structure of amphiphilic polymers enables them to maintain good viscosity and rheological properties in high salt environments, thus effectively maintaining the stability of emulsions [8].

Despite significant progress in research on the emulsification and stability of amphiphilic polymers, their practical application still faces many challenges. The emulsification ability and stability of specific salt-resistant amphiphiles need to be explore, and the effect of surfactants on the emulsification of amphiphiles is not clear. In this paper, the stability of type BIII amphiphilic polymer concentration on emulsion is investigated, and the effect of surfactant on its stability is also explored.

Experimental part

Experimental materials

Amphiphilic polymer: BIII amphiphilic polymer (an acrylamide-based amphiphilic polymer containing hydrophobic groups)

Water for solution preparation: 950 mg/L NaCl solution;

Crude oil: Daqing crude oil.

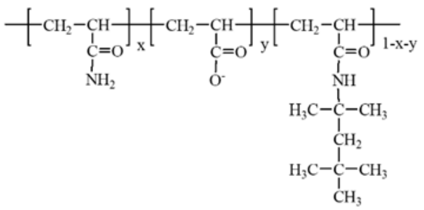

Figure 1. Molecular structural formula

Experimental apparatus

(1) Electronic balance, Sartorius Scientific Instruments (Beijing) Co;

(2) IKA RW200 Digital stirrer, Shanghai Renhe Scientific Instruments Co;

(3) FYL-YS-138L1 thermostat, Beijing Fuyi Co;

(4) Multi-head magnetic heating stirrer, Changzhou Guohua Instrument Co;

(5) Dispersing emulsifier FM200, Fluke Fluid Machinery Manufacturing Co;

(6) Digital display constant temperature water bath, Shanghai Meixiang Instrument Co;

(7) Brookfield DV-II+ Rotational Viscometer, product of Brookfield, USA;

(8) XSJ-2 type optical microscope, manufactured by Chongqing Optical Instrument Factory, China;

(9) TURBISCAN LAb Expert stability analyzer, ALV-GmbH, Germany.

Experimental methods

(1) Configuration of the target solution

Clearwater (mineralization degree 950 mg/L NaCl solution) was prepared with a concentration

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of 5000 mg/L mother liquor, and sewage water (mineralization degree 4000 mg/L NaCl solution) was used to prepare the destination solution. 5000 mg/L of BIII polymer and 950 mg/L of NaCl were introduced into 1 L distilled water and stirred using an IKA RW200 Digital stirrer. The initial rotation speed was 300 rpm and then was changed to 150 rpm after the amphiphilic polymer was evenly dispersed. 4–6 h later, the aqueous of dissolved amphiphilic polymer was put into a FYL-YS-138L1 thermostat at 45 °C for 24 h. 4000 mg/L NaCl was added into 1 L of distilled water to prepare the effluent.

200 ml aqueous with the mass concentration of 200 mg/L, 400 mg/L, 600 mg/L, 800 mg/L, 1000 mg/L and 1200 mg/L were prepared separately with the destination solution according to the ratio.

Add the required amount of mother liquor and sewage into a conical flask, and after stirring for 4–6 h with a multi-head magnetic heating stirrer, the flask was put into the FYL-YS-138L1 thermostat at a constant temperature of 45 °C to ripen for 24 h. The mixture of Fuyu crude oil and amphiphilic polymer was then prepared with the water in a conical flask.

The residual crude oil was mixed with the amphiphilic polymer solution according to the water-oil ratio of 1:3,1:2,1:1,2:1,3:1. Then the simulated emulsion was prepared by using a FM200 high-speed shear dispersing emulsifier with the rotational speed of 5000 rpm, the emulsification time of 15 min, and the emulsification temperature of 45 °C.

(2) Take microphotographs of the emulsion

Use a XSJ-2 optical microscope to take microscopic emulsion image and take different sizes on the photographs to measure the same example.

(3) Measurement of stability parameters

TURBISCAN Lab Expert type stability analyzer mainly analyzes the optical dispersion characteristics of the fluid by the pulse near-infrared light source. The transmittance and reflectance of light are different for different fluids or the same fluid with different stability at different times. The Stability Analyzer detects transmitted light and backscattered light through two detectors, and its curve can reflect the particle size growth or migration of emulsified oil droplets.

The intensity of the back scattered light BS is approximately inversely proportional to the square root of the photon transmission mean free range λ^* (1):

$$BS \approx \frac{1}{\sqrt{\lambda^*}} \quad (1)$$

The relationship between Transmitted light T and λ^* (Lambert-Beer law) (2–3):

$$T(\lambda *, r_i) = T_0\, e^{-\frac{2r_i}{\lambda *}} \quad (2)$$

$$\lambda^*(\varphi, d) = \frac{2d}{3\varphi(1-g)Q_s} \quad (3)$$

where r_i is the radius of the test chamber, T is the intensity of transmitted light in the continuous phase, D is the average particle size and φ is the particle volume concentration.

The stability parameter TSI is the stability analyzer for the data processing software used to evaluate the stability of the system (4):

$$TSI = \sqrt{\frac{\sum_{i=1}{}^{n}(x_i - x_{BS})^2}{n-1}} \quad (4)$$

where r_i is the average value of each backscattered light intensity and n is the number of scans. The smaller the TSI value is, the better stability the test system is.

Results and Discussion

Micrographs of emulsions with different concentrations of amphiphilic polymers and emulsion stabilization parameters

The curve of backscattered light intensity versus time is shown in Fig. 3. During the destabilization of emulsion, the decrease in the backscattered light intensity indicates the growth of the particle size of the emulsified oil droplets in the emulsion system at the initial stage which enlarged the particle size of emulsified oil droplets, resulting in the uplift

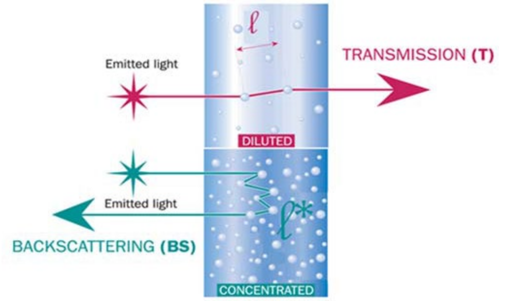

Figure 2. Stability analyzer working principal diagram

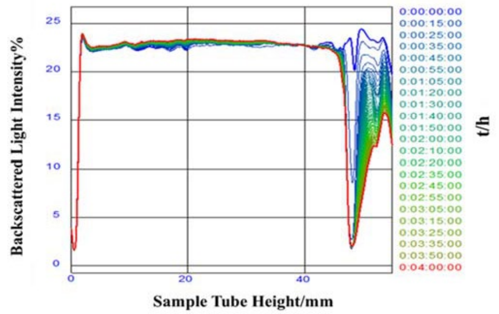

Figure 3. Multiple light scattering curves of emulsion with 1600 mg/L water-oil ratio 1:1

of oil droplets and the reduction of volume fraction of the dispersed phase at the bottom of the emulsion, thus leading to a decrease of backscattered light intensity. The changes in other ratios and polymer concentrations can be calculated with TSI values using similar curves.

Fig. 4 exhibited the influence of different oil-water ratio on stability of emulsion under at amphiphilic polymer concentration of 200mg/L. The effect of the parameter TSI in 4h measurement results shows that the smaller value of TSI indicated the more stable emulsion. The mixture with a ratio of Fuyu crude oil to amphiphilic polymer at 1:3 has the smallest TSI value, indicating that the formed emulsion is most stable, while the largest TSI value at the ratio of oil: polymer=1:3 indicates the worst stability.

Fig. 5 shows the emulsion stability at different oil-water ratios with a fixed amphiphilic polymer concentration of 800 mg/L. The TSI value is the smallest at a ratio of 1:3 of the amphiphilic polymer mixture to the residual crude oil, which means that the formed emulsion is more stable, while the water-oil ratio of 1:1 is improved compared to that of 200 mg/L.

Fig. 6 shows the stability of emulsions at different oil/water ratios for a certain amphiphilic polymer concentration of 1200 mg/L. The higher TSI for the 3:1 ratio indicates that the emulsion is unstable, but the TSI value stabilizes over time. The 1:3 ratio still shows the best stability, with little increase in TSI, and the 1:1 ratio is intermediate in terms of stability, with a relatively smooth TSI curve. At the polymer concentration of 1200 mg/L, the emulsion with a high water/oil ratio (3:1) shows lower stability than that with a lower water/oil ratio.

Fig. 7 at a fixed amphiphilic polymer concentration of 2000 mg/L gives the stability of emulsions at different oil-to-water ratios. At this high concentration, the 3:1 water-to-oil ratio still leads to the highest TSI indicating poor stability of emulsion while the 1:3 water-to-oil ratio maintains the best stability of emulsion with a lower TSI value.

Effect of particle size

The micrographs of emulsions with different concentrations are shown in Fig. 8. With the increasing concentration of amphiphilic polymer BIII, the particle size of droplets in the emulsion becomes smaller gradually and more uniformly dispersed, indicating the higher stability of the emulsion.

The larger the concentration of amphiphilic polymer, the smaller the average particle size of the emulsified oil droplets in the emulsion. The more uniform the particle size distribution, the more stable the emulsion is, which is consistent with the experimental results [11]. This can be attributed to that the amphiphilic polymer with higher concentration leads to the smaller particle size

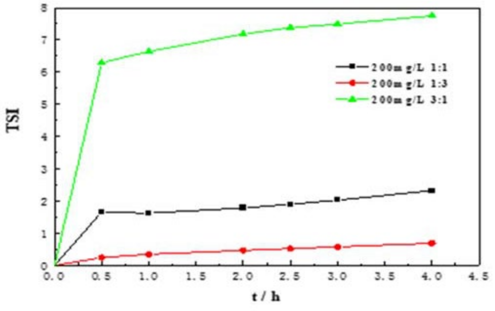

Figure 4. Polymer BIII concentration 200 mg/L emulsion

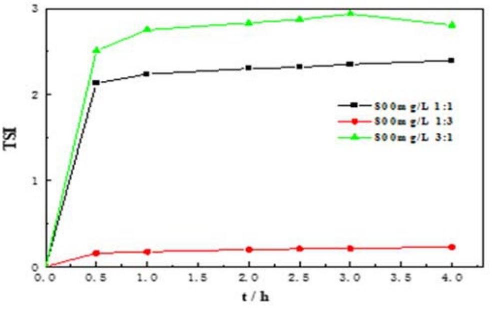

Figure 5. Polymer BIII concentration 800 mg/L emulsion

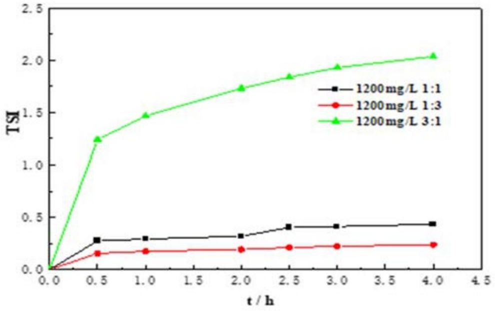

Figure 6. Polymer BIII concentration 1200 mg/L emulsion

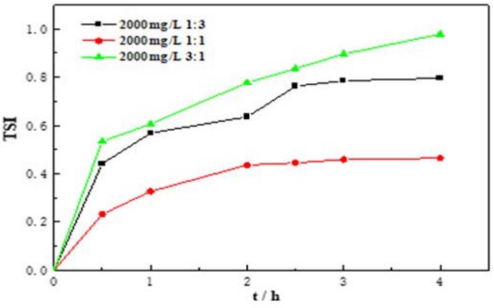

Figure 7. Polymer BIII concentration 2000 mg/L emulsion

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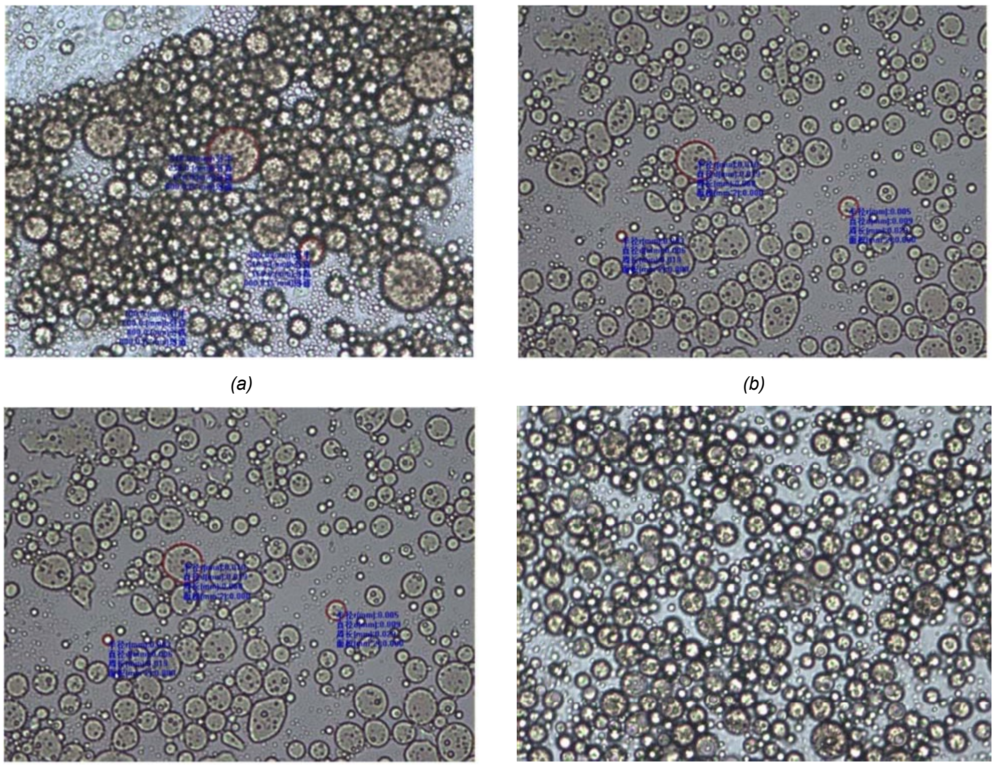

(a) *(b)*

(c) *(d)*

Figure 8. Polymer BIII concentration 2000 mg/L emulsion

(a) Polymer BIII concentration 200 mg/L; (b) Polymer BIII concentration 800 mg/L; (c) Polymer BIII concentration 1200mg/L; (d) Polymer BIII concentration 2000mg/L

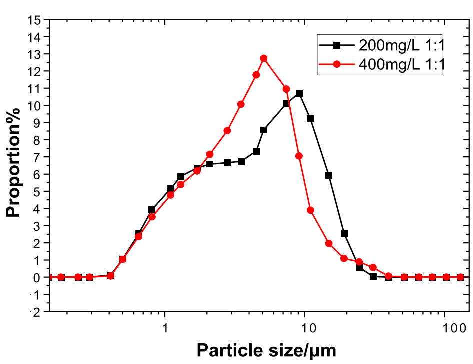

Figure 9. Particle size distribution

of emulsified oil droplets. The stronger the repulsion between droplets, the less likely to flocculation and aggregation, and thus the greater stability of the emulsion.

As the polymer concentration increases, the emulsion stability increases. The effect of amphiphilic polymers on the properties of the emulsion is reflected. The uplift rate of emulsified oil droplets in emulsion is shown in equation (5):

$$v = \frac{1}{18}[\frac{(\rho_w - \rho_o)gd^2}{\eta} \quad (5)$$

where v is the uplift rate of emulsified oil droplets, ρ_o is the density of the dispersed phase, ρ_w is the density of the dispersing medium, g is the gravitational acceleration, d is the diameter of the emulsified oil droplets, and η is the viscosity of the dispersing medium.

The uplift rate of emulsified oil droplets in the emulsion is related to the diameter of emulsion droplets and the viscosity of the dispersing medium, i.e., the higher the viscosity of the dispersing medium, the smaller the particle size of the emulsion droplets, and then the stability of the emulsion is elevated [12]. So it can be concluded that the effect of amphiphilic polymers on the apparent viscosity of the emulsion viscosity increases with the concentration of amphiphilic polymers increasing, therefore, high concentrations of amphiphilic polymers can enhance the viscosity of the emulsion and thus improve the stability of the emulsion. This can be attributed to the following properties of polymer: critical aggregation concentration. The amphiphilic polymers due to their molecular backbone with a small number of hydrophobic groups, will have intramolecular aggregation and intermolecular aggregation occurring when the concentration of amphiphilic polymers in the solution exceeds the critical aggregation concentration, thus forming a three-dimensional network structure in the solution. This phenomenon can cause the increasing hydrodynamic volume of the solution and an increase in the solution viscosity, which is different to these polymers, increasing the viscosity by increasing the molecular mass [13]. Some experiments have shown that at lower concentrations, the viscosity of aqueous solutions of hydrophobically bound polymers increases slowly with concentration, and when the critical concentration is exceeded, the viscosity of the solution tends to increase steeply and at a much higher rate than that of HPAM [14]. The effect of salinity on the amphiphilic polymer aqueous solution also have two effects. On the one hand, the electrolyte in the aqueous solution compresses the polymer diffusion bilayer structure, resulting in a decrease in the electrostatic repulsion between polymer molecules, which leads to the amphiphilic polymer long molecular chain curling contraction, thus resulting in a reduction of the apparent viscosity [10]. On the other hand, the addition of electrolyte will cause the polarity of the solution to increase, and the hydrophobic groups of polymer molecular chain will increase by increasing the interactions with each other, which will result in a decrease in the viscosity [15]. Through experimental observation, it is evident that the hydrophobically-conjugated water-soluble polymer aqueous solution shows insensitivity to salt, so it has good salt resistance. General water-soluble polymers due to their large relative molecular mass and long molecular chain, under the action of shear, will be its long molecular chain sheared under shear action, thus causing by a sharp decline in viscosity [3]. However, for amphiphilic polymers, due to the contained hydrophobic groups in the molecular chain, there are intramolecular and intermolecular associations in the solution, in the lower shear rate, the intramolecular associations to intermolecular associations occur in the transition, the apparent viscosity of the solution increases while, in the higher shear rate, the spatial network formed by intermolecular associations is destroyed, resulting in the decrease of the viscosity of amphiphilic polymers in the solution. Nevertheless, after the elimination of the shear effect, the solution viscosity can be restored to the original maximum level. However, after the elimination of the shear effect, the viscosity of the solution can be restored to the original highest level which is ascribed to that the hydrophobic groups on the polymer molecules re-form the structure of the bond [16].

With the further development of oilfields, the exploitation of high temperature and high mineralization reservoirs has gradually become the focus, but HPAM will be degraded at high temperatures and the viscosity will be greatly reduced under the high mineralization conditions, and it will inevitably be subjected to shear when it is used in the field, and the shear will result in the irreversible degradation of HPAM, which also affects the viscosity of its solution [5]. Amphiphilic polymers can be a good solution to the above problems, which not only helps to improve crude oil recovery in oilfields but also makes up for the shortcomings of ordinary polymer repellents that cannot be carried out in high-temperature and high-mineralization reservoirs in the past.

Conclusion

Emulsion stability enhance with the increase of amphiphilic polymer concentration. The increasing amphiphilic polymer concentration of the emulsion leads to the smaller average size of the emulsified oil droplets and more uniform particle size distribution, thus a stable emulsion. This can be attributed to the stronger repulsive effect between droplets which prevents the flocculation and polymerization of droplets, thus forming a more stable emulsion

The concentration of amphiphilic polymer has a greater impact on the emulsification of crude oil. If the concentration is lower than the CAC value, the formation of emulsion stability is poor, which is due to the main intramolecular aggregation of the amphiphilic polymer molecular chains in solution at this time while the aggregation of large molecular chains resulting in emulsion stability is greatly enhanced when the amphiphilic polymer concentration is higher than the CAC value. The results is consistent with the results of the experimental analysis.

Mechanism of emulsifying crude oil by amphiphilic polymers can be ascribed to the presence of hydrophobic groups.

ADDITIONAL INFORMATION

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