Increasing Effect of Water Clarifiers on the Treatment of Polymer-Containing Oil Production Sewage

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Residual anionic polyacrylamide in polymer-flooding oil production wastewater results in the formation of a thermodynamically stable system. In this study, the effects of three different types of medicaments, namely, cationic, anionic and nonionic agents, in dynamic treatments, such as adding a position, dosage and combined processes of chemical addition, on the oil removal rate of sewage were examined. In the treatment with a single agent, the oil removal rate of the cationic agent CQY-1 and the nonionic agent CHF-2 was ≥ 97.8%. The charge characteristics of different ionic agents for the combined dosing treatment indicated that the oil removal rate was better than that of a single agent; the combined dosing ratio was 50 mg/L CHP-1 and 50 mg/L CHP-2. At 80 mg/L COY-1, the oil removal rate of the dynamic process was ≥ 98.8%, and the dosage of CQY-1 was reduced from 200 mg/L to 50–150 mg/L, which corresponded to a decrease of 25.0%–75.0%. Therefore, the combined dosing process effectively reduced the single dosage.

Keywords: cationic agent; increasing effect; oil removal rate; water clarifiers.

INTRODUCTION

Polymer-flooding oil recovery is an important technical means to increase and stabilize production in oil fields. Polymer injection significantly improves crude oil recovery. A polymer injected into the formation undergoes shear, degrades and returns with the produced fluid in the form of low-molecular-weight anionic polyacrylamide. Several residual polymers exist in this type of polymer-containing sewage1,2, thereby causing difficulties in removing oil, increasing the amount of degreasing water agent, decreasing the oil removal rate and yielding a poor water quality effect3,4.

However returning from production wastewater causes several common problems in polymer flooding oilfield; these problems include increased sewage viscosity, enhanced electronegativity, minimized oil aggregation and collision and deteriorated gravity sedimentation effect5–7. During offshore oilfield exploitation, the offshore platform has a short processing time and limited space, so a quick oil removal treatment is a critical step1. The addition of a water-clearing agent can significantly improve the quality of sewage discharged through crude oil processing. The type of agent must be adjusted, the adaptability of the agent with the existing process must be explored, and the changing emulsification of the produced water must be adapted8–11. The efficiency of various increasing oil removal processes has been examined to effectively treat polymer-injected oily sewage, ensure the normal operation of water treatment and achieve water quality standards.

Here, an evaluation method for a clearing agent with a small amount of drug added at different stages in the process was established to efficiently treat polymer-oily sewage. The oil removal rate of different kinds of agents in the process was explored in the experimental treatment process according to the field test data of the process design. The oil removal effects of three cationic, anionic and nonionic types of water agents were investigated under a high oil removal rate, a low dosage of chemicals, a low concentration of medicament and a stable process by using different concentrations of wastewater as the experimental object. The influences of the increasing effect, the position of the added chemicals, the optimized combination and the amount of the three types of water clarifiers were also examined. The results indicated that the increasing amounts of water clarifiers and the dosing process showed a broad application prospect in the treatment of polymer-containing oilfield wastewater.

MATERIALS AND METHODS

Experimental instruments and chemicals

For wastewater containing polymer and oil, simulated samples were prepared by the sewage collected from an oil field, and their salinity was 9581.62 mg/L. The amounts of polymer and oil were 180.0 and 800.0–4300.0 mg/L, respectively, according to the actual measurement. The zeta potential value was ζ = −23.17 mV. The names of the water clarifiers are shown in Table 1. The water clarifiers used in the experiment were cationic (COY-1, COY-2, COY-3, COY-4 and COY-5), anionic (CHP-1, CHP-2 and CHP-3) and nonionic (CHF-1 and CHF-2) types. Medicament was utilized for the low zeta potential of different ionic agents and sewage. A chemical reaction quickly occurred to break the thermodynamic equilibrium system and achieve oil bead aggregation and separation.

The following experimental instruments were used. A dynamic flow evaluation device of the oil and water treatment agent with convenient visualization and disassembly was established based on marine sewage treatment. The following devices were also utilized: a sewage heating mixing tank (V = 18 L), inclined plate deoiling devices (inclination angle of the inclined plate is 45°), a gas flotation device, a walnut shell-quartz sand dual media filter (DN60 × 600 mm) and other assisted instruments that could be used alone or combined into
different experimental units. The supporting equipment also involved a FLUKO FM25 high-shear dispersing emulsifier and a SHIMADZU 2450 UV–visible spectrophotometer.

**EXPERIMENTAL METHOD**

The device (Fig. 1) was convenient to carry and could be used for the on-site evaluation of oil-water treatment chemicals in the laboratory and the platform. Each unit of the process could be disassembled and loaded into two suitcases. The dimensions (Length × Width × Height) of the suitcases were 763 mm × 483 mm × 300 mm (inclined plate deoiling devices) and 585 mm × 442 mm × 300 mm (gas flotation device). The simulated sewage containing polymer and oil entered the inclined plate deoiling devices once. Then, the sewage entered the gas flotation device to remove suspended solids and oil and proceeded to the walnut shell filter for filtration. The experimental procedure had four continuous dosing points (A–D) and four sampling points (I–IV). The following process control parameters were set: the inclined plate treatment residence time was 15–25 min, the gas flotation treatment dwell time was 8–18 min, and the running time of the entire process of the sewage referred to the on-site process setting and could control 20–45 min.

The prepared sewage containing polymer and oil was added to the sewage buffer tank and mixed for 8–10 min. The constant temperature of the sewage buffer tank was 60°C. After the pump displacement was set, it was started, and polymer–oil wastewater was added to each unit. The water clarifiers were added based on the predesigned dosing points. After the sewage was filled with the whole instrument, the samples were taken at four sampling points along the process, and the oil content was measured. The dynamic flow parameters of each processing unit were set as follows: 23 min of stay time of the inclined plate, 280 mL/min sewage pump displacement, air flotation intake gas of 340 mL/min and air flotation sewage residence time of 15 min.

The final concentration of the residual oil in each sample was determined using petroleum ether as the oil-extraction solvent in accordance with the oil concentration-absorbance standard curve equation for examining water and wastewater:

\[ m_1 = 555.56A_1 + 1 \]  

where \( m_1 \) is the oil content (mg/L), and \( A_1 \) is the measured absorbance. The absorbance of the sample was analyzed at a wavelength of 410–430 nm with a spectrophotometer.

**RESULTS AND DISCUSSION**

**Increasing effect for combined dosing process**

Three types of chemical agents with a high oil removal rate were selected by comparing the oil removal experiment and combination dosing processes.

#1: Nonionic water clarifier + cationic water clarifier (CHF-2 + CQY-1),
#2: Nonionic water clarifier + anionic water clarifier (CHF-2 + CHP-1 + CHP-2),
#3: Anionic water clarifier + cationic water clarifier (CHP-1 + CHP-2 + CQY-1).

**Increasing effect for cationic agent**

The zeta potential of the polymer-containing sewage was negative (\( \zeta = -23.17 \text{ mV} \)). The dosage of the agent was depended on the charge characteristics of different ion agents and the oil content of the sewage.

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**Table 1. Types and components of water clarifiers**

| Types          | Drug name and Structure composition                          | Name   |
|----------------|---------------------------------------------------------------|--------|
| Cationic clarifier | Polydimethylallyl ammonium chloride (PDMDAAC) \( \cdot \) (A.R) | CQY-1  |
|                 | Cationic polyacrylamide (relative molecular mass of 5 million) \( \cdot \) (A.R) | CQY-2  |
|                 | Cationic polyacrylamide (relative molecular mass of 10 million) \( \cdot \) (A.R) | CQY-3  |
| Nonionic clarifier (Laboratory preparation) | A multi-branched nonionic surfactant having a molecular structure of \( \text{D(PO)}_3\text{(EO)}_y\text{(PO)}_z\text{H} \). Where: EO-polyoxyethylene; PO-polyoxypropylene; R-fatty alcohol; D-polyethylene polyamine; x, y, z-degree of polymerization; | CHF-1  |
|                 | The main component is polyoxyethylene polyoxypropylene stearyl ether, the theoretical structural formula is: \( \text{R(PO)}_x\text{(EO)}_y\text{(PO)}_z\text{H} \). Where: EO-polyoxyethylene, PO-polyoxypropylene, R-fatty alcohol; x, y, z-degree of polymerization; | CHF-2  |
| Anionic clarifier | \( \text{ZnCl}_2 \) 60%, \( \cdot \) (A.R) | CHP-1  |
|                 | Anionic polyacrylamide (relative molecular mass of 10 million) + \( \text{ZnCl}_2 \) 60%, \( \cdot \) (A.R) | CHP-2  |
|                 | Anionic polyacrylamide (relative molecular mass of 15 million) + \( \text{ZnCl}_2 \) 60%, \( \cdot \) (A.R) | CHP-3  |

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**Figure 1.** Experimental process for dynamic evaluation of sewage treatment agent performance
Different concentrations of CQY-1, CQY-2, CQY-3, CQY-4 and CQY-5 were added at point B. Explore the influence of different chemicals on the degreasing rate of sewage (the original water samples in Figures 2a–e below were marked with #1).

Figure 2a shows that the oil content in water gradually increased during CQY-1 treatment after different amounts of chemicals were added. The increasing effect heightened as the agent concentration increased. When the dosage was 200 mg/L, the increasing oil rate of the entire process reached 97.8%, thereby showing an excellent effect.

Figures 2b and c show that the trend of the oil concentration caused by the change in the amount of the agents was the same as in CQY-2 and CQY-3, and the oil removal rate of the process was 94.5%. The oil content of the sewage increased, and the oil removal effect improved. However, the difference between both factors was obscured. At sampling point I, the increasing effect of the high concentration of the water clarifiers was better than that of with a low concentration. The oil content of the outlet of the inclined plate deoiling devices was the same. The results indicated that the increasing effect of the two agents in the entire process was constant even though the speed of action increased as the dosage increased. Thus, the appropriate dosing amount was 150 mg/L.

Figure 2d demonstrates that the reaction of CQY-4 was relatively slow. At sampling point I, the oil content of the inclined plate inlet was basically equal. After the inclined plate deoiling devices were reached, the effect on the reaction was obvious. The oil removal effect of the high dosage was better than that of the low dosage, and the difference in the effect after air flotation was minimal. When 250 mg/L CQY-4 was added, the inclined plate deoiling devices had basically no increasing effect. The agent mainly relied on the gas flotation device to use a large number of microbubbles and gather the oil droplets and suspended matter with small particle size and remove oil. The oil removal rate reached 63.2%, but the increasing rate of the entire process was slightly lower than that at a dosage of 350 mg/L. This observation could be attributed to the presence of the agent in the inclined plate deoiling devices after the dosage increased, the subsequent relied on the air flotation devices and the double media filter to remove oil, and the increasing effect of each unit could be reflected. CQY-4 slightly changed the oil removal rate of the entire process when the dosage was 250–350 mg/L. Figure S1e shows that the increasing effect of the sewage improved, and the oil removal rate of the process was 93.5% when the amount of CQY-5 was 350 mg/L.

Figures 2a–e demonstrate that the oil concentration of the inclined plate deoiling device outlet (point II) was low. The oil removal rate was more than 85.5% at the outlet of the inclined plate deoiling devices. The oil and flocs significantly increased after they reached the gas flotation device, thereby indicating that the agent quickly reacted in the pipeline. The oil concentration was reduced by more than 54.1%. The oil was further removed by the inclined plate deoiling devices where the agents and the sewage continuously reacted. The inclined plate deoiling devices quickly coalesced.
and settled to remove most of the oil. Consequently, the concentration of sewage oil greatly decreased, thereby minimizing the pressure of the subsequent process along the path. A cationic agent was added between the sewage tank and the inclined plate deoiling devices to obtain an improved increasing effect. The comparison of the dynamic experimental results of the five types of cationic agents revealed that the increasing rate at the dosing point B reached 97.5% when CQY-1 was optimal at 200 mg/L.

**Increasing effect for nonionic agents**

CHF-1 and CHF-2 were added at 100 and 150 mg/L at point A to observe the effects of different chemicals on the oil removal rate of the sewage (Fig. 3).

The nonionic agent was directly added to the sewage tank. Thereafter, oil floating in the sewage tank was observed. The oil beads in the inlet of the inclined plate with several oil slicks were quickly collected. The oil and flocs significantly decreased after they reached the gas flotation device. The oil content of sewage could be reduced to less than 8.6 mg/L. Figure 3 illustrates that the oil removal rate of the inclined plate outlet was 31.9% higher than that of the inclined plate inlet when the dosage of CHF-1 was 100 mg/L. CHF-1 significantly increased the oil removal rate of the entire process by 9.6% (from 100 mg/L to 150 mg/L). CHF-2 showed an excellent oil removal effect (≥93.7%, point I) on the sewage tank even before the inclined plate. The oil removal rate of the entire process was 98.4% (point IV) when the dosage was 100 mg/L. However, increasing the amount of the agent had no obvious effect on the improvement of the oil removal rate.

![Figure 3](image-url)

**Figure 3.** Changes in oil content along the process when adding nonionic agents

The addition of CHF-1 and CHF-2 at the dosing point A could significantly reduce the oil content of the sewage, and the oil removal effect of CHF-2 was optimal. When the dosage was 100 mg/L, the oil content of the sewage at the entrance of the inclined plate was reduced to 56.5 mg/L. The agent quickly reacted, and the oil-water separation effect was remarkable. CHF-2 is a type of water clarifier that mainly promotes the mutual aggregation of oil beads by changing the oil-water interface. This agent also removes certain fine solid particles that are often adsorbed on the surface of oil beads during the increasing process. CHF-2 is compatible with anionic polymers in production liquid. The point of adding the nonionic water clarifier was arranged before the oil-water separation of the wastewater. The effect of the water clarifier could be fully observed because of the large number of oil beads in the sewage tank and the probability of collision with one another. The accumulation of oil beads could be accelerated to reduce difficulties in further sewage treatment. Therefore, CHF-2 could be used as a rapid coalescing agent when it was combined with other agents.

**Increasing effect for anionic agent**

CHP-1 was combined with CHP-2 or CHP-3 as an anionic water-clearing agent. Points B and C were the points of the addition of the agent on the outlet pipe of the sewage pump. A micromixer was installed between points B and C to enhance the reaction effect of the agent. In the experiment, the relationship between the order of CHP-2/CSP-3 and CHP-1 and the effect of sewage treatment was evaluated to analyze the change in oil contents.

Figure 4 shows that the combined effect of CHP-2 and CHP-1 was better than that of CHP-3 and CHP-1, and the oil removal rate of the entire process was high. The combination of CHP-2 and CHP-1 was added to the pipeline before the inlet of the inclined plate inclined deoiler. The dosing sequence was as follows: CHP-2 was added at point B, and CHP-1 was added at point C. After sampling was conducted at point II, the increasing effects of CHP-2 and CHP-1 were better than those of CHP-1 and CHP-2. The finding indicated that this method mainly relied on the inclined plate, air flotation and double medium filtration to remove oil. The increasing effect was mainly reflected on the inclined plate unit. Therefore, the dosing order of CHP-1 (point B) and CHP-2 (point C) was good. The increasing effect of the CHP-3 and CHP-1 combination on the inclined plate unit was poor. In this case, the addition of 150 mg/L to the CHP-1 (point B) + CHP-2 (point C) anionic agent had an optimal increasing effect with an oil removal rate of ≥95.1%.

![Figure 4](image-url)

**Figure 4.** Changes in oil content along the process when adding anionic agents

**Increasing effect for combination dosing process #1**

The combined dosing process #1 involved CHF-2 (point A) + CQY-1 (point B). Cationic agents have an excellent increasing ability in oilfield wastewater treat-
ment, display the fast flocculation rate and flocculation onset time and flocs are viscous and easy to aggregate. As such, an on-site process is performed to reduce the number of cationic agents and efficiently remove oil. In the present study, the amount of the cationic agent was reduced, and the change in the increasing effect was tested. The different dosages of #1 were as follows: 

- **a:** 80 mg/L CHF-2 + 150 mg/L CQY-1,
- **b:** 60 mg/L CHF-2 + 150 mg/L CQY-1,
- **c:** 50 mg/L CHF-2 + 180 mg/L CQY-1,
- **d:** 80 mg/L CHF-2 + 60 mg/L CQY-1,
- **e:** 80 mg/L CHF-2 + 100 mg/L CQY-1.

Figure 5 shows that the oil content of the sewage at sampling point I rapidly decreased to 24.7–26.3 mg/L when the dosages were **a, b** and **c**. The oil in water was substantially removed when **d** and **e** were applied. In the addition of **a** and **b**, the increasing effect was improved when the CHF-2 dosage was reduced, and the CQY-1 dosage was unchanged. This finding indicated that the cationic agent still plays a leading role in the increasing effect. The result of the comparison between the single agent and the added **d** and **e** (Fig. 6) demonstrated that the concentration of CQY-1 remarkably influenced the increasing effect of CHF-2. At sampling point I, when CHF-2 and CQY-1 were 80 and ≥150 mg/L, respectively, the increasing rate was higher than that of CHF-2. This result indicated that the latter could promote the increasing effect of the former. When CQY-1 was ≤100 mg/L (such as **d** and **e**), the increasing rate was significantly reduced, and CQY-1 had a certain inhibitory effect on the increase in CHF-2. The inclined plate deoiling devices were used to effectively reduce the oil content of the sewage and keep a high oil removal rate in sampling point II.

The oil content in the sewage in the entire process decreased when the dosage of the agent in #1 was changed. The dosages of the nonionic and cationic agents decreased by 20.0% and 60.0%, respectively, and the oil removal rate of the entire process was ≥96.4%. In this combination, CHF-2 had an optimal increasing effect when it was added to the sewage buffer tank, thereby decreasing the number of cationic agents. CHF-2 destroyed the membrane properties of the oil-water interface and promoted the effective aggregation between the oil droplets and the suspended particles. CQY-1 was applied to compress and destroy the double electric layer through an adsorption bridging action, to achieve the accumulation of oil droplets and suspended solids and to form large floating flocs for clearing water and remove oil. A combination effect was better than the increasing effect of the single agent, and this effect could be obtained by adjusting the amount of CHF-2 and CQY-1 to be added. When dosage a was added to the combined agent, the increasing effect at the sampling point I was better than that of the single agent. Nevertheless, the oil concentration slightly fluctuated along the process possibly because of the mutual influence of the two agents.

**Increasing effect for combination dosing process #2**

The combined dosing process #2 involved CHF-2 (point A) + CHP-1 (point B) + CHP-2 (point C). The different dosages of #2 were as follows: 

- **f:** 100 mg/L CHF-2 + 150 mg/L CHP-1 + 150 mg/L CHP-2,
- **g:** 50 mg/L CHF-2 + 100 mg/L CHP-1 + 100 mg/L CHP-2,
- **h:** 50 mg/L CHF-2 + 50 mg/L CHP-1 + 100 mg/L CHP-2,
- **i:** 100 mg/L CHF-2 + 100 mg/L CHP-1 + 50 mg/L CHP-2.

The amounts of CHF-2, CHP-1 and CHP-2 were reduced, the oil removal effect of the process unit equipment was constant, and the oil removal rate of the sewage was still ≥99.5%. Thus, the oil removal effect of the combination of the agents was better than that of the single agents (Fig. 7). The dosage of the agent was reduced by 33.3%, and the oil removal rate reached 99.5%. The dosage of CHF-2 was decreased by 50.0%, and the dosages of CHP-1 and CHP-2 were decreased by 66.7 and 33.3%, respectively. The oil removal rate of the entire process was still ≥95.4%. When CHP-1 and CHP-2 were used alone, the oil removal rate of the inclined plate deoiling devices was less than 86.4% (Fig. 8). The addition of CHF-2 reduced the number of various water clarifiers and significantly diminished the oil content in water.

**Increasing effect for combination dosing process #3**

The combined dosing process #3 included CHP-1 (point B) + CHP-2 (point C) + CQY-1 (point D). In #3, the dosage of the agent was changed on the basis of...
the additional amount of $j$. The oil removal rate of the process and the oil content in the entire process were studied when the dosage of the agent decreased. The different dosages of #3 were shown as follows:

- $j$: 100 mg/L CHP-1 + 100 mg/L CHP-2 + 150 mg/L CQY-1,
- $k$: 50 mg/L CHP-1 + 50 mg/L CHP-2 + 80 mg/L CQY-1,
- $m$: 80 mg/L CHP-1 + 80 mg/L CHP-2 + 100 mg/L CQY-1,
- $n$: 80 mg/L CHP-1 + 60 mg/L CHP-2 + 80 mg/L CQY-1,
- $p$: 80 mg/L CHP-1 + 80 mg/L CHP-2 + 50 mg/L CQY-1.

**CONCLUSIONS**

CQY-1 (200 mg/L) was added at point B, CHF-2 (100 mg/L) was added at point A, and CHP-1 (points B) and CHP-2 (points C) were added, respectively. The dosage was 150 mg/L, and the oil removal rate of the entire process was $\geq 97.8\%$. The oil removal efficiency of the combined dosing process was better than that of the single agent. This process could effectively reduce the amount of the cationic agent. The amount of CQY-1 to be added was reduced from 200 mg/L to 50–150 mg/L, which was approximately 25%–75%. The oil removal rate of the inclined plate unit was $\geq 96.7\%$.

Figure 9 illustrates that the combined agent effect was superior to the single-agent effect. After the addition of an agent, the reaction rate rapidly increased. When the dosage was $j$, the oil removal rate of sampling point I reached 96.2\%. A few oil slicks were observed on the surface of the inclined plate deoiling and gas flotation devices, and the viscosity was weak. When 80 mg/L CHP-1, 80 mg/L CHP-2 and 100 mg/L CQY-1 were added, the optimal oil removal effect was obtained. The oil should be removed by the inclined plate settler when $k$ and $p$ were added (Fig. 10). Figure 10 also shows that the oil removal rate at sampling point I decreased as the concentration of the CQY-1 agent decreased in combined dosing. The addition of any agent affected the oil removal rate of sampling point I. The effects of the anionic and cationic agents were observed in the combined test of CHP-1, CHP-2 and CQY-1. The dosages of the anionic and cationic agents decreased by 66.7 and 46.7\%, respectively, and the oil removal rate of the entire process was $\geq 98.8\%$. CHP-1 and CHP-2 promoted oil droplet aggregation and coalescence whilst avoiding electrical interaction with the polymer. Without changing the sewage treatment process, the increasing effect of the combined agents was better than that of the single agent. Therefore, the dosage of the combined agents was greatly reduced compared with that of the single agent. A similar phenomenon was noticed in other studies.13–15.
The inclined plate unit and the oil removal rate of the entire process were both 98.8%.

The results of this experiment can be used to optimize the dosing process and dosing amount of polymer-containing sewage treatment on the oilfield site, to reduce the dosage of chemicals and improve the oil removal rate of sewage and provides a new solution for the oil removal of polymer-containing sewage.

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