

Research Article

# Viscoelastic Oil Displacement System of Modified Silica Nanoparticles/Zwitterionic Surfactant for High Salinity Reservoir

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Polymer flooding and polymer/surfactant flooding have achieved good efficiency in the application of conventional reservoir, but the existed chemical flooding technology cannot address the issues of the requirements of chemical flooding in high salinity reservoir. Under the condition of high salinity reservoir, due to the increase of calcium and magnesium ions, the increasing viscosity effect of oil displacement system is lost. In order to study the feasibility of applying nanomaterials in the field of enhanced oil recovery under the conditions of high salinity reservoir, develop a low-concentration and high-efficiency oil displacement system. EAPC solution has advantages in reducing interfacial tension, but its viscosity is not good. Therefore, hydrophobically modified silica nanoparticles (SiO<sub>2</sub> NPs) were added to the carboxylic acid-type erucic acid amide propyl betaine (EAPC) solution. The interaction between EAPC and hydrophobic carbon chains led to the exposure of carboxyl groups, thus making the system more stable. The interfacial activity and zeta potential were studied, and the interaction mechanism between modified SiO<sub>2</sub> NPs and EAPC was obtained. The results show that when the EAPC concentration is 0.3%, the apparent viscosity of the modified silica nanoparticles (SiO<sub>2</sub> NPs) composite system can reach 40 mPa·s, and the oil-water interfacial tension can be reduced to 10<sup>-2</sup> mN/m. The micro-visualization model and the simulated oil displacement experiment proved that the modified SiO<sub>2</sub> NPs (0.3%)/EAPC (0.3%) composite system has a variety of oil displacement mechanisms. Under the simulated reservoir conditions (total salinity of 25000 mg/L, calcium and magnesium ion concentration of 500 mg/L, 70 °C), it is proved that the modified SiO<sub>2</sub> NPs composite system had good viscoelasticity and improved oil washing efficiency. The oil displacement system has guiding significance for effectively enhancing the recovery of high salinity reservoir.

## 1. Introduction

Due to the increasing global demand for energy, the development of cost-effective enhanced oil recovery (EOR) technologies is an important part of meeting future energy needs [1]. Surfactant flooding can use crude oil that cannot be recovered only by water flooding, effectively reducing the remaining oil saturation in reservoir and playing an important role in tertiary oil recovery. With the development of many oil fields entering the middle and late stages,

the available resources of conventional reservoirs are becoming less and less, and it is more difficult to further enhance oil recovery [2, 3].

In recent years, the research direction has gradually shifted to the development of unconventional reservoirs such as and high salinity [4, 5]. Due to the characteristics of high salinity, higher requirements are put forward for surfactant for oil displacement. A new type of viscoelastic surfactant can be used in high temperature and high salinity reservoirs. It can not only improve oil washing efficiency

by reducing oil-water interfacial tension, but also play a role in improving sweep efficiency similar to polymer with high viscosity. It also has the advantages of temperature resistance and salt tolerance and has broad application prospects in improving oil recovery. The previous research have reported that betaine-based amphoteric surfactant based on erucic acid structure have strong viscosity-increasing properties. It can form wormlike micelles when compounded with surfactants or polymers to enhance viscoelasticity [6–9].

Because of their special physical and chemical properties, silica nanoparticles ( $\text{SiO}_2$  NPs) have been used in many industrial fields [10, 11]. Many researchers have focused on the study of the interaction between  $\text{SiO}_2$  NPs and surfactant and their synergistic effects on interfacial properties [12, 13]. Wang et al. [14] studied on spheric  $\text{SiO}_2$  nanoparticles with the sizes range in 10–100 nm which are dispersed in non-aqueous solvent under the assistant of surfactant. When the volume fraction of surfactant is 6%, it shows good dispersion. However, the high surface energy of  $\text{SiO}_2$  NPs seriously affects their stability and dispersibility in aqueous solutions, which limits their wide application. Chemical surface modification is a common method to improve the stability of  $\text{SiO}_2$  NPs in their aqueous dispersions. Wang et al. [15] studied the influence of silica nanoparticles on surface properties, especially the interaction between surfactant and particles. Zhang et al. [16] studied poor temperature resistance of fracturing fluid in high temperature, high salinity, and low permeability reservoirs. By adding silica nanoparticles, the strength and stability of cross-linked structure were increased, and the temperature resistance of fracturing fluid was improved. The surface-modified  $\text{SiO}_2$  NPs will have special surface properties due to the reduction of the number of hydroxyl groups and the introduction of new functional groups, which are more complex to interact with surfactant, and there are fewer studies related to them [17].

Therefore, in view of the characteristics of high salinity and strong heterogeneity reservoir, this paper aims to investigate the effects of hydrophobically modified  $\text{SiO}_2$  NPs on the interfacial tension, viscosity, and rheological properties of the amphoteric surfactant erucamidopropyl betaine, and a high-efficiency viscoelastic surfactant system was constructed through the hydrophobic interaction between them. The results of the oil displacement experiments showed that the system has good oil displacement performance under the condition of high salinity reservoir.

## 2. Experimental Procedures

**2.1. Materials.** The erucamide propyl carboxylic betaine (EAPC) was synthesized according to the literature [8]. The molecular structure of the resulting sample is shown in Figure 1, and the purity was confirmed by  $^1\text{H}$ NMR spectrum. 1-hexadecanol (98 wt%), p-toluenesulfonic acid monohydrate (98 wt%), ethanol ( $\geq 99.5$  wt%), toluene (98 wt%), n-octane (98 wt%), sodium chloride (NaCl), calcium chloride ( $\text{CaCl}_2$ ), and magnesium chloride ( $\text{MgCl}_2$ ) (all analytical purity) were purchased from Sinopharm Group. The hydrophilic  $\text{SiO}_2$  NPs was purchased from Aladdin Reagent Com-

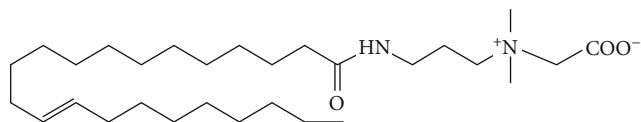


FIGURE 1: The molecular structures of EAPC.

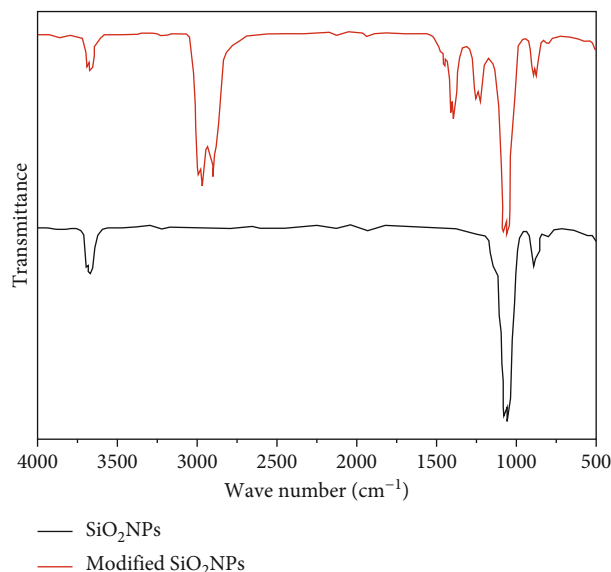


FIGURE 2: FTIR spectra of unmodified and modified  $\text{SiO}_2$  NPs.

pany, with a particle size of 7 nm~40 nm and a surface area of  $200 \text{ nm}^2/\text{g}$ . All materials were used as is.

The experimental water was brine prepared with sodium chloride, with a concentration of 25000 mg/L. The experimental oil was the dehydrated crude oil from Bohai SZ36-1 Oilfield.

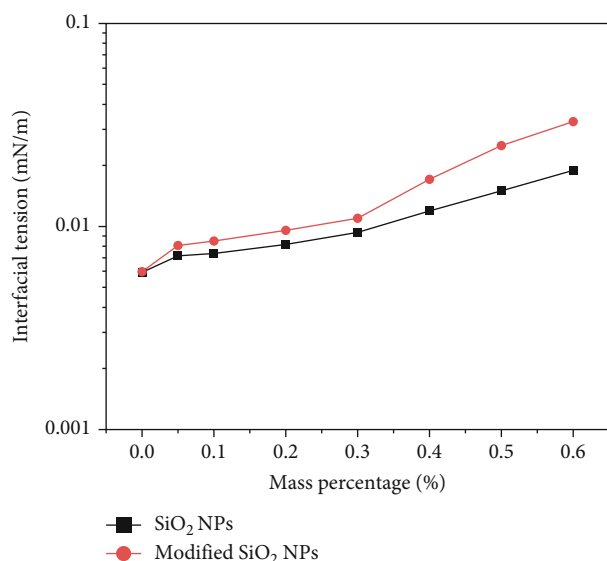
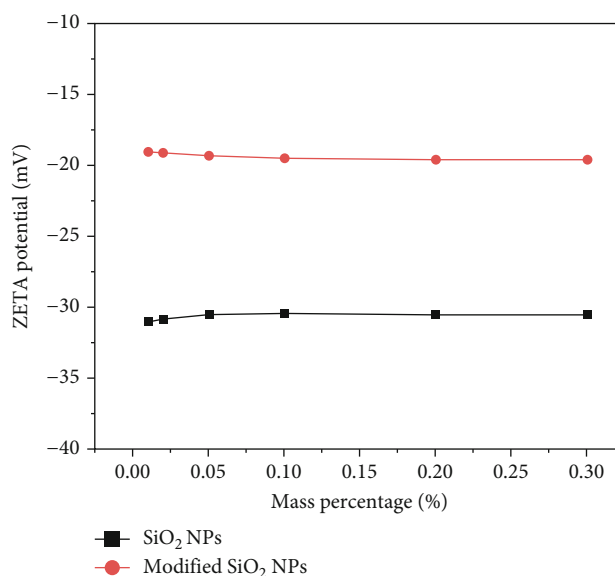
**2.2. Modification and Characterization of  $\text{SiO}_2$  NPs.** In a three-necked flask, 5 g  $\text{SiO}_2$  NPs, 12.5 g 1-hexadecanol, and 0.2 g p-toluenesulfonic acid monohydrate were added to 300 mL of toluene. The mixture was then heated to  $120^\circ\text{C}$  and kept under reflux and stirring for 4 h. After the reaction, a white solid was obtained by vacuum-rotary evaporation, which was further washed 3 times with ethanol. Finally, the product was dried in a vacuum oven at  $100^\circ\text{C}$  for 24 h to obtain modified hydrophobic  $\text{SiO}_2$  NPs.

In order to determine the functional groups of  $\text{SiO}_2$  NPs, the Fourier transform infrared spectroscopy (FTIR) spectra of the samples were tested by Nicolet iS50 FTIR (American Thermo Nicolet, Inc.).

**2.3. Experimental Methods for the Performance of Composite Systems.** The oil-water interfacial tension was determined using a TX-500 spin-drop interfacial tension meter according to relevant standards for the given temperature conditions. A Malvern Zetasizer Nano ZSP instrument (Malvern Company) was used to measure the zeta potential of individual NP dispersion and surfactant/NPs dispersion. The apparent viscosity and rheological properties of the

TABLE 1: The changes of viscosity and interfacial tension of EPAC at different concentrations.

Concentration (%)	0.1	0.2	0.3	0.4	0.5
Viscosity (mPa-s)	4.8	5.3	5.6	5.9	6.1
Interfacial tension (mN·m <sup>-1</sup> )	0.057	0.026	0.0063	0.0057	0.0042

FIGURE 3: Effects of mass fraction of unmodified and modified SiO<sub>2</sub> NPs on the IFT between EAPC solution (0.3%) and crude oil.FIGURE 4: Zeta potential changes of unmodified and modified SiO<sub>2</sub> NPs (0.1%) with the increased concentration of EAPC.

experimental system were tested by Brookfield LVDV-III viscometer and Physica MCR 302 rheometer.

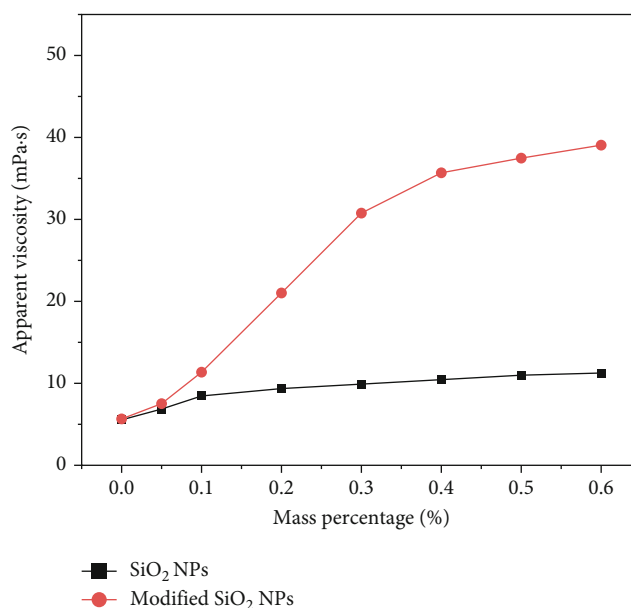
**2.4. Microscopic Visualization of Oil Displacement Experiments.** The microscopic glass pore model was evacuated and saturated with brine, and then crude oil was injected into the model at a rate of 0.5 mL/min. After the saturation of oil was completed, the brine and the chemical flooding system are injected successively at the same speed to observe the displacement characteristics and the remaining oil distribution.

**2.5. Simulated Core Oil Displacement Experiments.** The oil displacement effect of the system was evaluated by the DHZ-50-180 chemical flooding state simulation device at a temperature of 70 °C. Artificial cores were used during flooding experiments [18, 19]. After water flooding to water cut of 98% at the outlet end, water flooding oil recovery was calculated. Then inject 0.5 PV (PV is the pore volume) of the oil displacement agent slug, and following water flooding until the water cut at the outlet end is 98%, and the final oil recovery is calculated.

### 3. Experimental Results Analysis

#### 3.1. Effect of Nanomaterials on Properties of EAPC Solution

**3.1.1. FTIR Spectra of Nanomaterials.** The FTIR spectra of unmodified hydrophilic and modified hydrophobic SiO<sub>2</sub> NPs are shown in Figure 2. It can be seen from the figure that the absorption peaks at 900 cm<sup>-1</sup>, 1000-1100 cm<sup>-1</sup>, and

FIGURE 5: Effects of mass fraction of unmodified and modified SiO<sub>2</sub> NPs on the viscosity of EAPC solution (0.3%).

3400 cm<sup>-1</sup> in both samples correspond to the bending vibration of Si-OH and the stretching vibration of Si-O-Si and -OH, respectively. It was found that the -OH (3400 cm<sup>-1</sup>)

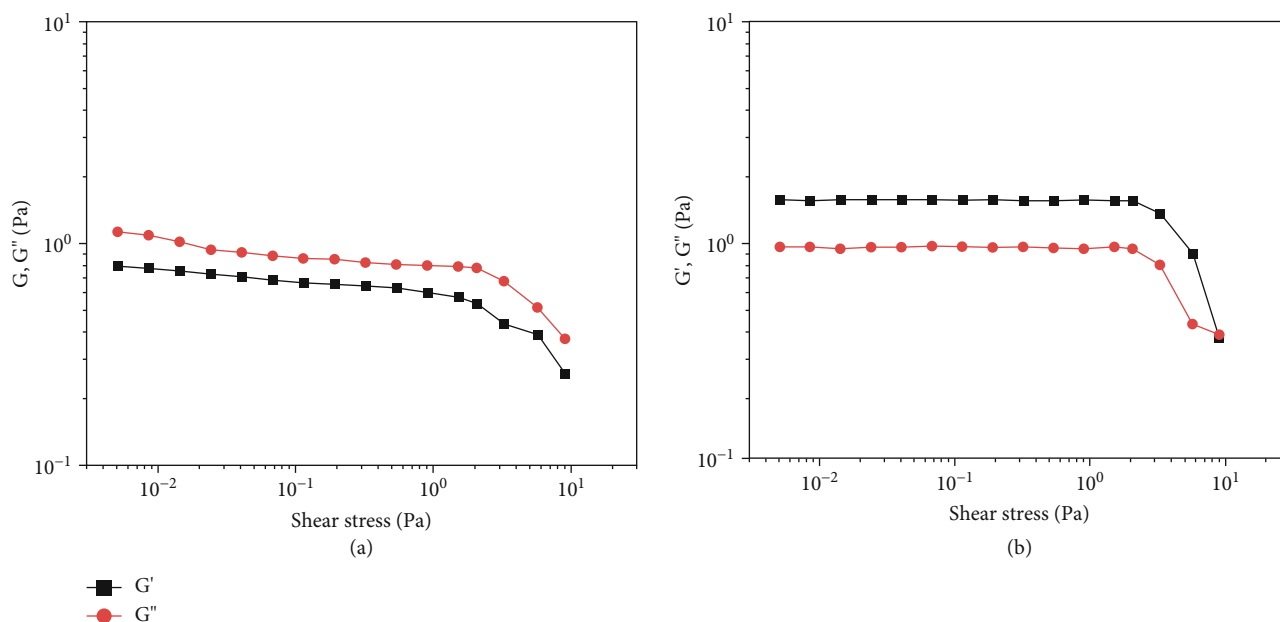


FIGURE 6: The curves of storage modulus ( $G'$ ) and loss modulus ( $G''$ ) with shear rate for the mixed NPs/EAPC system (0.3 wt%). (a) unmodified  $\text{SiO}_2$  NPs/EAPC. (b) modified  $\text{SiO}_2$  NPs/EAPC.

absorption intensity of the modified  $\text{SiO}_2$  NPs was significantly weakened, which was caused by the reduction of the surface hydroxyl groups of the  $\text{SiO}_2$  NPs. New absorption peaks appear at  $2850\text{-}2960\text{ cm}^{-1}$ ,  $1460\text{ cm}^{-1}$ ,  $1300\text{ cm}^{-1}$  and  $1200\text{ cm}^{-1}$ , corresponding to  $-\text{CH}$ ,  $-\text{CH}_3$  ( $-\text{CH}_2-$ ),  $\text{CO}$ , and  $\text{CC}$ , respectively, indicating that 1 cetyl alcohol molecules have been successfully grafted on the surface of hydrophilic  $\text{SiO}_2$  NPs [20].

**3.1.2. The Properties of EPAC.** It can be seen from Table 1 that when simulated the high salinity reservoir NaCl concentration is  $25000\text{ mg/L}$ , in the experimental concentration range, the viscosity of EPAC was between  $4\text{ mPa}\cdot\text{s}$  and  $6\text{ mPa}\cdot\text{s}$ , the change was very small, and the viscosity increase ability is weak. With the increased of concentration, the oil-water interfacial tension gradually decreased. When the concentration exceeded 0.3%, the interfacial tension value could reach the order of magnitude of  $10^{-3}\text{ mN/m}$  and indicated that EPAS had a good effect on reducing the oil-water interfacial tension.

**3.1.3. The Influence of Nanomaterials on the Interfacial Tension of EAPC Solution.** The synthesized carboxylic acid type erucamidopropyl betaine (EAPC) can reduce the water/crude oil IFT to less than  $10^{-2}\text{ mN/m}$  at lower concentration (0.3%), but the solution viscosity is relatively low ( $5.6\text{ mPa}\cdot\text{s}$ ). We added the unmodified and modified  $\text{SiO}_2$  NPs to the 0.3% EAPC solution to investigate their influence on the interfacial tension of the surfactant. The results are shown in Figure 3. The surface of  $\text{SiO}_2$  NPs is negatively charged, and when the positively charged nitrogen atoms inside the EAPC molecule are attracted by the negative electricity, the molecule will approach the surface of the nanoparticle laterally. At this time, the carboxyl group at the

end of the molecule will be in contact with the surface repulsive force, which affects the adsorption of EAPC molecules on the surface. Therefore, the addition of hydrophilic nano-silica has a limited effect on the arrangement of EAPC in the interface adsorption layer. For the modified  $\text{SiO}_2$  NPs, the number of surface hydroxyl groups is reduced, and there are hydrophobic chains composed of sixteen carbon atoms distributed, which causes more EAPC molecules to be adsorbed to the surface of NPs through hydrophobic interactions, thereby reducing the surface activity in the oil-water interface layer. The effective concentration of the agent molecule causes an increase in IFT, and the value is higher than that of the system where hydrophilic  $\text{SiO}_2$  NPs are added.

In order to investigate the interaction between EAPC and modified  $\text{SiO}_2$  NPs, the zeta potential changes of unmodified modified and  $\text{SiO}_2$  NPs in EAPC solution were compared, as shown in Figure 4. From the figure, it showed that with the increase of EAPC concentration, the zeta potentials of the two NPs dispersions did not change significantly. This is related to the fact that the amphoteric surfactants do not show electrical properties to the outside, resulting in a phenomenon similar to the adsorption of non-ionic surfactants. However, the zeta potential of the two NPs showed different trends. For the unmodified NPs, the zeta potential showed a slight increase, while the modified NPs showed the opposite change. Since the surface of  $\text{SiO}_2$  NPs is negatively charged, it will attract the positively charged part of EAPC and make it adsorb laterally on the surface. The exposed carboxylic acid group is affected by the adjacent positively charged N atoms, and the negative charge is weakened. Therefore, the overall zeta potential increases with the adsorption of EAPC. For the modified NPs, the adsorption of EAPC relies on the interaction

between the hydrophobic carbon chains, and the carboxyl group is exposed to the outside, which causes the zeta potential to rise slightly.

**3.2. Effect of Modified  $\text{SiO}_2$  NPs on Rheological Properties of EAPC Solution.** The effects of different mass fraction of unmodified and modified  $\text{SiO}_2$  NPs on the viscosity of EAPC solution (0.3%) is shown in Figure 5. From the figure, it can be seen that compared with the unmodified NPs, the modified  $\text{SiO}_2$  NPs had a significant impact on the viscosity of the EAPC solution and significantly increased the apparent viscosity of the EAPC solution. Due to the nature of amphoteric surfactant, there are worm-like micelles inside the 0.3% EAPC solution, but the length is short and not enough to form a stable spatial structure. The viscosity of the solution is only 5.6 mPa·s. There are hydroxyl groups existed on the surface of unmodified  $\text{SiO}_2$  NPs. In addition to the electrostatic effect, the hydrogen bond between the nanodispersion and the EAPC micelles promotes the spatial structure of the association in the solution to a certain extent, causing the viscosity of the solution to rise slightly. The hydrophobic interaction between the long carbon chains on the surface of the modified  $\text{SiO}_2$  NPs and the micelles has an adhesion effect, which enhances the mutual attraction between the weaker micelles, promotes the strength of the association structure, and causes the viscosity of the solution to increase significantly and makes the EAPC aqueous solution viscoelasticity.

In order to research the dynamic viscoelasticity of the system, it is necessary to determine the linear viscoelastic region of the system. Fix the frequency ( $f = 1 \text{ Hz}$ ), and obtain the storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of the system with shear stress. The area where the modulus does not vary with the shear stress is the linear viscoelastic zone. The change curve of the storage modulus and loss modulus with the shear rate of the unmodified or modified  $\text{SiO}_2$  NPs and EAPC mixed system with a mass fraction of 0.3% is shown in Figure 6. As shown in Figure 6(a), in the system with unmodified  $\text{SiO}_2$  NPs, the loss modulus is greater than the storage modulus from the beginning, and both moduli tended to decrease with the rise of shear stress, indicating that the solution has no viscoelasticity. For the system with modified  $\text{SiO}_2$  NPs, the two moduli remain basically unchanged before the stress reaches a certain value and start to decrease. The initial state of the elastic properties of the system is greater than the viscous properties, which is due to the formation of the network structure of the worm-like micelles in the system.

From Figure 6(b), the stress value  $\sigma = 0.8 \text{ Pa}$  is selected in the linear viscoelastic region, and the frequency sweep of the above system is performed to obtain the relationship between the modulus and the frequency (Figure 7). As shown in the figure, the loss modulus of the system is higher than the storage modulus in the low frequency range and lower than the energy storage modulus at higher frequency. Most human worm-like micelles exhibit this property, which conforms to the Maxwell model and indicates that the system possesses a certain degree of viscoelasticity [21].

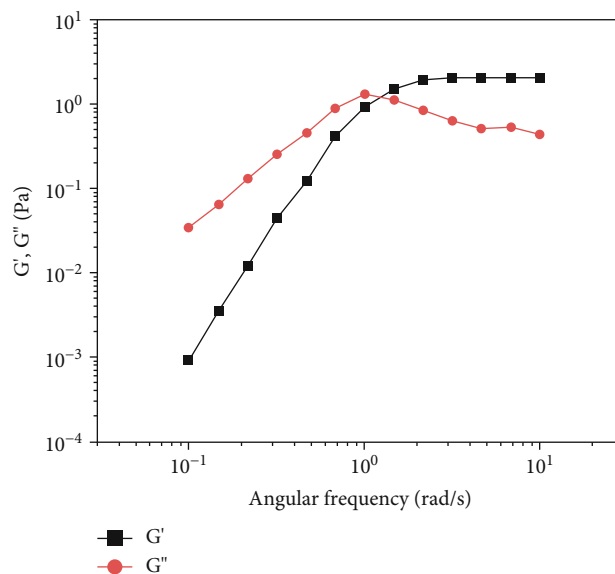


FIGURE 7: The curves of storage modulus ( $G'$ ) and loss modulus ( $G''$ ) with angular frequency for the mixed NPs/EAPC system (0.3 wt%).

**3.3. Study on Microscopic Oil Displacement Performance of Modified  $\text{SiO}_2$  NPs/EAPC Composite System.** At the end of chemical flooding, the oil displacement effect of the modified  $\text{SiO}_2$  NPs/EAPC composite system (0.3%  $\text{SiO}_2$  NPs and 0.3% EAPC) was studied through the microscopic visualization oil displacement model, and the unmodified  $\text{SiO}_2$  NPs/EAPC solution was selected as a control. The experimental results are shown in Figures 8 and 9. Obviously, the composite system with modified  $\text{SiO}_2$  NPs has good interfacial tension and higher viscosity and has good oil displacement efficiency and larger affected area within the same injection time. It can be seen from Figure 9 that the modified  $\text{SiO}_2$  NPs can make the EAPC solution more effectively displace the oil film on the pore wall and the residual oil at the blind end, which is closely related to its viscoelasticity. The normal stress and shear stress of the viscoelastic fluid have a stronger carrying effect on the oil film, and extrusion swelling effect can increase the mobility of the residual oil at the blind end and effectively reduce the residual oil saturation [22].

**3.4. Study on Macroscopic Oil Displacement Performance of Modified  $\text{SiO}_2$  NPs/EAPC Composite System.** Use the unmodified and modified  $\text{SiO}_2$  NPs composite system (0.3%  $\text{SiO}_2$  NPs and 0.3% EAPC) to conduct indoor simulation oil displacement experiments to verify the performance of the viscoelastic composite system to improve oil recovery. The summary of the core flooding tests using the mixed  $\text{SiO}_2$  NPs/EAPC system is shown in Table 2.

In the simulated reservoir (salinity of 25000 mg/L, calcium and magnesium concentration of 500 mg/L, 70 °C), the modified NPs composite system can effectively increase the crude oil recovery rate by 20.1% (Table 2), which is significantly higher than that of the original system with modified NPs added (7.9%). The unmodified NPs cannot

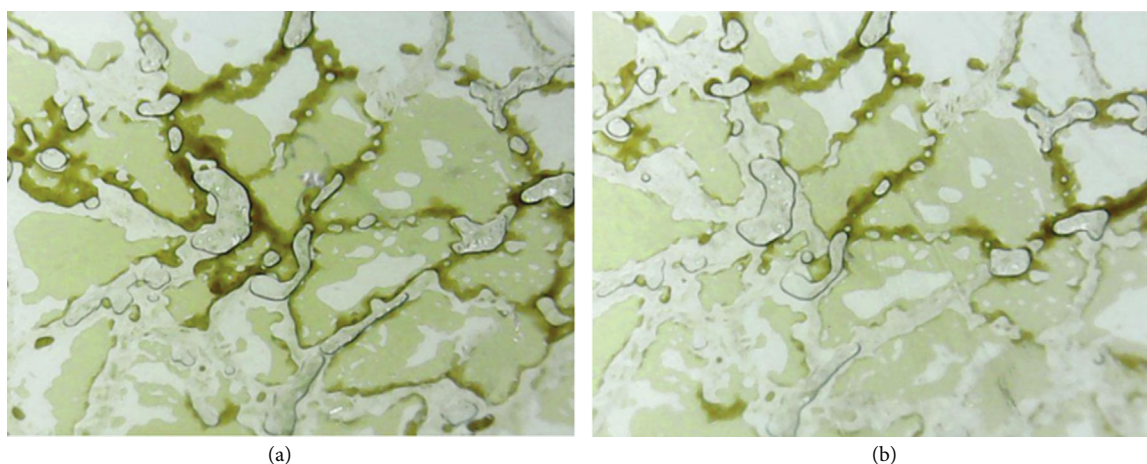


FIGURE 8: Two displacing results in microscopic visual model: (a) unmodified  $\text{SiO}_2$  NPs/EAPC; (b) modified  $\text{SiO}_2$  NPs/EAPC.

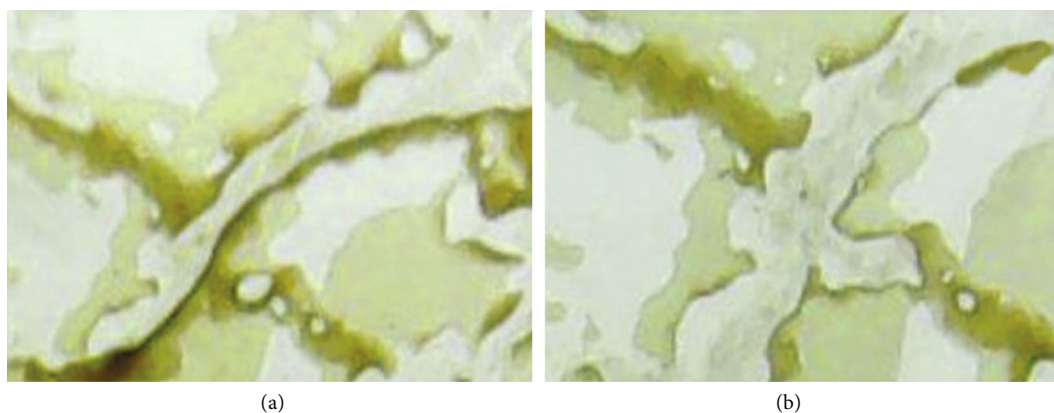


FIGURE 9: Local residual oil distribution: (a) unmodified  $\text{SiO}_2$  NPs/EAPC; (b) modified  $\text{SiO}_2$  NPs/EAPC.

TABLE 2: Summary of the core flooding tests using the  $\text{SiO}_2$  NPs/EAPC composite system.

Oil displacement system	Gas permeability (mD)	Water flooding recovery rate (%)	Final recovery rate (%)	Improving recovery rates (%)
0.3% EAPC+0.3% unmodified NPs	1085	54.3	62.2	7.9
0.3% EAPC+0.3% modified NPs	1130	58.1	78.2	20.1

produce a synergistic effect with EAPC, so it can only rely on EAPC to reduce IFT and slightly increase viscosity to improve oil recovery, and the effect is very limited. The composite system added with modified  $\text{SiO}_2$  NPs relies on the nanoparticle dispersion grafted with long hydrophobic chains for the overlap between EAPC micelles, which is less affected by the external environment and not only maintains a certain viscosity under severe conditions, but also increases the sweep coefficient. At the same time, it also has the oil displacement mechanism of viscoelastic surfactant, which has better mobilization of oil film and residual oil at the blind end, and improves the efficiency of oil displacement.

#### 4. Conclusion

By adding hydrophobically modified  $\text{SiO}_2$  nanoparticles (NPs) to 0.3% carboxylic acid erucamidopropyl betaine

(EAPC) solution, a composite system with certain viscoelasticity was constructed.

- (1) By comparing with unmodified  $\text{SiO}_2$  NPs, the results of interfacial tension and zeta potential show that EAPC relies on the hydrophobic interaction between carbon chains to adsorb on modified  $\text{SiO}_2$  NPs
- (2) The modified  $\text{SiO}_2$  NPs relies on the hydrophobic effect to overlap the short EAPC micelles to form a spatial structure in the solution, produce a viscosity-increasing effect, and make the solution viscoelastic
- (3) The visual oil displacement model and core displacement experiment proved that the viscoelastic composite system has the effect of improving the oil displacement efficiency and expanding the swept volume, showing a good application prospect

## Data Availability

The all data included in the manuscript.

## Conflicts of Interest

The authors declare that they have no conflict of interest.

## Acknowledgments

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