

Research Article

Effect Evaluation of Nanosilica Particles on O/W Emulsion Properties

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O/W emulsion reinforced by nanosilica particle has good application in hydrocarbon development. However, there are few reports on the influence of nanosilica particles on the oil-water interface of O/W emulsion. The effect of nanosilica particles on the interfacial properties of O/W emulsion was indirectly investigated by measuring the interfacial properties between aqueous dispersion of nanosilica particles and kerosene, and the properties of O/W emulsion reinforced by nanosilica particle were studied. The results showed that the aqueous dispersion of nanosilica particles could significantly reduce the interface tension (with kerosene) by more than 50%, and the interface tension between the aqueous dispersion and kerosene decreased with the increase in nanosilica content. The aqueous dispersion of nanosilica particles could significantly change rock wettability. When the content of nanosilica particles increased from 0.1% to 0.7%, the contact angle decreased from 44.89° to 27.62°. The surface tension of O/W emulsion prepared by the aqueous dispersion of nanosilica particles and kerosene was among 25 mN/m~30 mN/m. The contact angle was also particularly small, with an average of about 20.00°, a minimum of 12.50°. The salts had little effect on the interface tension of emulsions but had a significant influence on the contact angle and its stability. Magnesium salt could reduce the three-phase contact angle and increase the hydrophilic properties of O/W emulsion, while calcium salt had the opposite effect. Calcium salt and magnesium salt could reduce the stability of the emulsion, and calcium salt had a greater influence. The oil-water stratification adding either calcium salt or magnesium salt was about 1 day~3 days earlier than that without salts. In the experiment, when the content of nanosilica particles was among 0.3%~0.7%, the viscosity of O/W emulsion increased with the increase in nanosilica particles. When the content was 0.9%, the viscosity suddenly decreased, and the extent of reduction was about 21.7%. The findings of this study can help for better understanding the application of nanosilica particles in O/W emulsion, giving some suggestions for the application of nanoparticles in hydrocarbon development.

1. Introduction

With the increasing depletion of hydrocarbon resources and the increasingly prominent contradiction between supply and demand, it has always been the hot issue to dig the development potential of the reservoir after water flooding and further improve both the utilization degree of the reservoir and the resource recovery in the field of hydrocarbon

resource development [1–4]. Therefore, various methods have been used in the attempt to improve oil recovery [5–9]. Fracturing and acidizing are two mainly recommended methods for reservoir stimulation, and the role of the stimulation techniques is to improve the seepage capacity of the reservoir and reduce the seepage resistance [10, 11]. For conventional low-permeability reservoirs, they can accelerate the production rate, and for ultra-low-permeability

reservoirs such as shale and tight sandstone reservoirs, it can help to improve the ultimate recovery [12–14]. The chemical flooding has always been the most commonly employed techniques for enhancing oil recovery [15–18], such as surfactant flooding, polymer flooding, alkali flooding, emulsion flooding, and various chemical combination flooding. The mechanism of enhancing oil recovery by chemical flooding has two aspects: increasing sweep efficiency and enhancing oil displacement efficiency. Besides, CO₂ flooding and foam flooding are also widely applied for enhancing oil recovery [19, 20] and their mechanism is similar to that of chemical flooding.

Among them, emulsion flooding is one of the most important methods to enhance oil recovery [21, 22] and shows a good application further in enhanced oil recovery after water flooding. Meanwhile, studies have shown that in situ emulsification of crude oil is also an important means to enhance oil recovery, especially for heavy oil reservoirs [23–26]. Besides, there are also reports on the application of emulsion combined with carbon dioxide or foam to enhance oil recovery [27, 28].

As an oil-water composite system, O/W emulsion has good application in both adjusting the flow profile of reservoir fluid and improving the fluid sweep coefficient. Besides, in recent years, with the development of nanotechnology and the integration of interdisciplinary approaches, nanotechnology has been gradually applied to various fields of reservoir development, showing good applications [29–32].

Nanotechnology is the design and application of engineered or naturally occurring nanoparticles with the order of 1–100 nm to accomplish specific purposes [33–35]. Nanoparticles possess two unique properties of ultrasmall particle size and ultra-high-specific surface area. And material properties are sized dependent at the nanoscale; therefore, nanoparticles can be engineered to contain specific optical, magnetic, interfacial, thermal, electrical, or chemical properties to perform specific functions. Combined together, these unique properties allow nanoparticles to be used for many purposes in the oilfield. Nanosilica, nanoalumina, nanotitanium dioxide, nanoferric oxide, and nanocopper oxide are all the nanomaterials used in the development of hydrocarbon resources [36–38], and they are served in various aspects such as drilling fluid filtration reduction [39, 40], cementing [41, 42], temperature sensor and thermal conductivity improvement [43], slow release of surfactant [44], tracer, foam, and emulsion stability improvement [45–49], fracturing fluid performance improvement [50, 51], EOR, and oily sewage treatment [37, 38, 52, 53], achieving good results. Besides, the retention and adsorption of nanoparticles in porous media and their influence on fluid transportation are also reported [54–59]. Most of the above reports on the application of nanoparticles in hydrocarbon development are about nanosilica particles. And nanosilica particles are widely favored in reservoir development due to their wide source and high cost performance.

In the reports on the emulsion stabilized by nanoparticles, it was mainly about the effect of emulsion on enhancing oil recovery with the synergies of nanoparticles. However, there were few reports especially on the effect of nanoparticles on the interfacial properties of O/W emulsion.

However, it is very important to study the effect of nanosilica particles on the oil-water interface properties of O/W emulsion for understanding the mechanism of nanosilica particles strengthening O/W emulsion, and it can provide reference for the application of nanoparticles in hydrocarbon development.

The previous reports were mostly about the application of nanosilica particles in oil and gas development, and there are no reports about the effect of nanosilica particles on oil-water interface properties of O/W emulsion. In the O/W emulsion system, nanosilica particles tend to adsorb on the oil-water interface due to the difference between oil and water, changing the interfacial properties, so as to change the properties of O/W emulsion.

Therefore, it is of great significance to study the effect of nanosilica particles on the oil-water interface of emulsion. However, there are no methods or means to directly determine the interfacial properties of O/W emulsion currently. In order to study the effect of nanosilica particles on O/W emulsion, the O/W emulsion interface reinforced by nanosilica particles is regarded as the interface between the aqueous solution of nanosilica particle and the oil phase. The oil-water interfacial properties of the O/W emulsion are approximately characterized by measuring the interfacial properties between the two phases. And the general sketch of the study is shown in Figure 1.

This method has some certain limitations. Since the homogeneous O/W emulsion reinforced by nanosilica particle is regarded as two phases—the aqueous solution of nanosilica particles and oil phase, this method can only qualitatively analyze the influence of nanosilica particles on the interfacial properties of O/W emulsion. It is necessary to explore other experimental methods to accurately describe the distribution of nanosilica particles in the O/W emulsion system and the influence of nanosilica particles on the oil-water interfacial properties in O/W emulsion. Besides, the study is only carried out with the temperature 30°C, without investigating the effect of other temperatures.

In the study, different concentrations of nanosilica aqueous solution and O/W emulsion reinforced by nanosilica were firstly prepared, respectively. Then, the surface tension of nanosilica aqueous solution, the interfacial tension between nanosilica aqueous solution and kerosene, and the contact angle of nanosilica aqueous solution were investigated with different concentrations of nanosilica. After that, the effects of nanosilica particles on the stability and viscosity of O/W emulsion were investigated. Finally, the effects of salt content on the stability of O/W emulsion reinforced by nanosilica and the properties of O/W emulsion reinforced by nanosilica in the presence of sodium ions and magnesium ions were also investigated. Totally, the experimental procedure can be summarized as shown in Figure 2.

2. Laboratory Experiments

2.1. Experimental Apparatus and Chemicals. Experimental apparatus mainly include an SFZL-A automatic surficial/interfacial tensiometer, balance, blender, HWY-10 thermostat

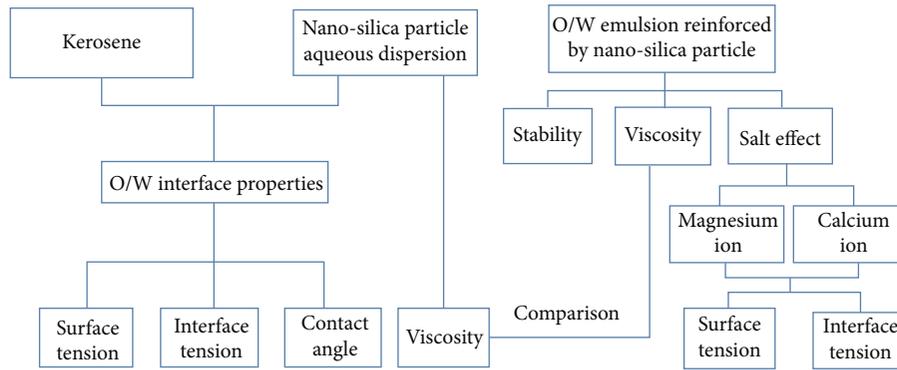


FIGURE 1: The general sketch of the study.

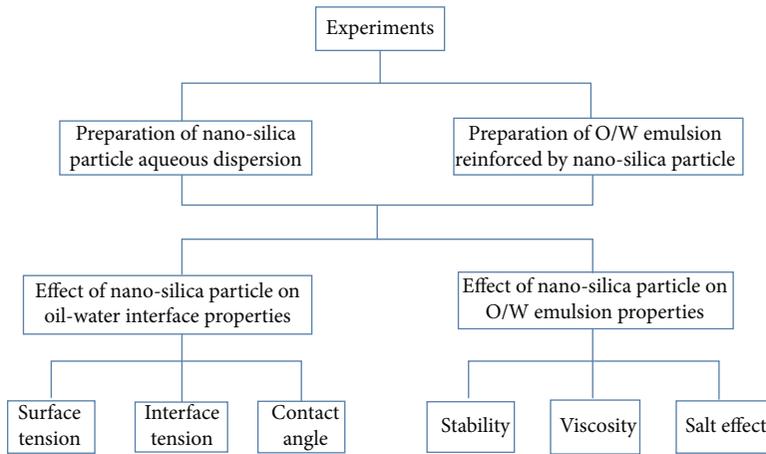


FIGURE 2: The flowchart of the experiments.

water bath, JGW-360a contact angle meter, rotary viscometer, etc.

The main chemicals used in the experiment are nanosilica particles (mean particle size 20 nm), emulsifier FC, distilled water, kerosene, calcium chloride, magnesium chloride, etc.

2.2. Experimental Methods

2.2.1. Preparation of O/W Emulsion. Firstly, nanosilica particles and emulsifier FC were added to the beaker, and then, the mixture was diluted with distilled water to the required concentration. The nanosilica particles and emulsifier were fully dispersed in distilled water by stirring for 5 minutes in a constant speed blender. Then, kerosene was added to the beaker and stirred at a high speed for 20 minutes in a constant speed blender to obtain O/W emulsion reinforced by nanosilica particles.

2.2.2. Effect of Nanosilica Particles on Oil-Water Interface Properties. The nanosilica particles were added into distilled water and fully stirred, so that the nanosilica particles were fully dispersed in distilled water, and the aqueous dispersion of nanosilica particles was obtained. The interface properties between nanosilica particle dispersion and kerosene were evaluated by using the SFZL-A automatic surficial/interfacial tensiometer and JGW-360a contact angle meter, and the

effect of nanosilica particles on the oil-water interface properties was quantified.

2.2.3. Effect of Nanosilica Particles on O/W Emulsion Properties. O/W emulsions reinforced by different contents of nanosilica particles were prepared, and the stability, rheological properties, and salt resistance of O/W emulsions with nanosilica particles were evaluated. The main instruments included the NDJ-85 rotary viscometer and HWY-10 thermostat water bath.

3. The Experiment Result Analysis

3.1. Effect Evaluation of Nanosilica Particles on Interface Properties. The oil-water interface has a direct impact on the emulsion properties; however, the oil-water interface properties of O/W emulsion cannot be directly measured and characterized. Interface tension and contact angle are two commonly recommended parameters to characterize the interface properties. So considering the adsorption of nanosilica particles on the oil-water interface, the interface tension and contact angle between the aqueous dispersion of nanosilica particles and kerosene were measured, and the effect of the nanosilica particle adsorption on the oil-water interface properties of the emulsion was indirectly quantified by interface tension and contact angle.

3.1.1. Surface Tension of Aqueous Dispersion with Nanosilica Particles. The aqueous dispersions of nanosilica particles with the contents of 0.1%, 0.3%, 0.5%, 0.7%, and 0.9% were prepared, respectively. The surface tension of aqueous dispersions with different contents of nanosilica particles was measured, and the surface tension of distilled water was used as a reference to clarify the effect of nanosilica particles on the surface tension of aqueous dispersion. In order to ensure the accuracy of the measurements, the surface tension was measured several times and their average was taken as the value of surface tension. The specific results are shown in Figure 3.

It could be obtained from Figure 3 that the surface tension of aqueous dispersion with nanosilica particles showed the explicit trend of declining with the increase in nanosilica particle content. When the content of nanosilica particles was among 0.1%~0.5%, the surface tension decreased rapidly. When the content of nanosilica particles was about 0.5%, the surface tension decreased with the increase in the nanoparticle contents. While the content of nanosilica particles was 0.9%, the surface tension reached the minimum, 54.33 mN/m.

The nanosilica particles in the aqueous dispersion tended to be gradually adsorbed on the surface of the dispersion, with the influence of the interfacial difference between the dispersion and the air, and the adsorption amount increased with the content of nanosilica particle increasing, thus reducing surface tension of the aqueous dispersion.

3.1.2. Interface Tension between Aqueous Dispersion of Nanosilica Particles and Kerosene. The aqueous dispersions of nanosilica particles with the contents of 0.1%, 0.3%, 0.5%, 0.7%, and 0.9% were prepared, respectively, and the interface tensions between aqueous dispersions with different contents of nanosilica particles and kerosene were measured.

The interface tension between water and kerosene was taken as the reference, and the specific results are shown in Figure 4. The interface tension between aqueous dispersion of nanosilica particles and kerosene decreased rapidly and fluctuated slightly with the increase in nanosilica particle content. Overall, there was little difference in interfacial tension between aqueous dispersion with different silica contents and kerosene, and the interface tension between them fluctuated in the range of 17 mN/m~19 mN/m, which was significantly less than that between kerosene and water (40.26 mN/m).

The nanosilica particles in aqueous dispersion were adsorbed on the oil-water interface due to the difference in properties at the oil-water interface, which played a role in reducing the interface tension. Moreover, the ability of nanosilica particles to reduce the interface tension was related to their adsorption capacity. As solid particles, the adsorption capacity of nanosilica particles on the oil-water interface was limited, so the adsorption capacity on the interface had reached saturation with small content of nanosilica particles. Therefore, the ability to reduce the interface tension no longer increased with the increase in nanosilica particle content.

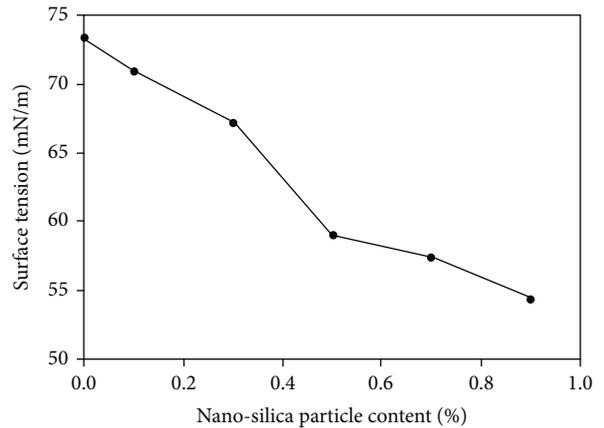


FIGURE 3: Relationship between nanosilica particle content and surface tension.

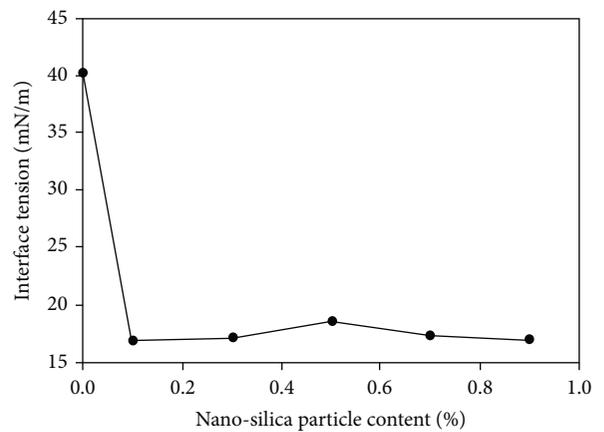


FIGURE 4: Relationship between nanosilica particle content and interface tension.

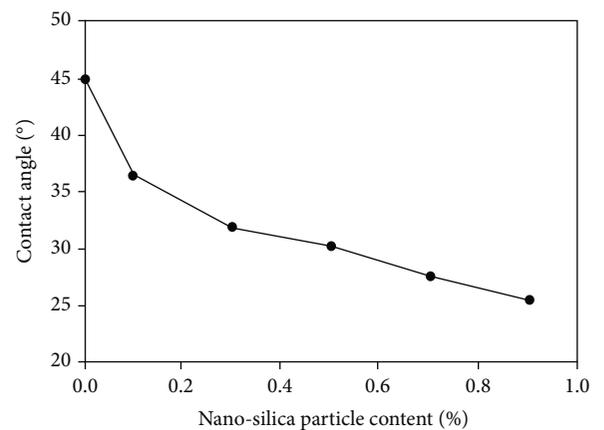


FIGURE 5: Relationship between nanosilica particle content and contact angle.

3.1.3. Contact Angle of Aqueous Dispersions with Nanosilica Particles. The contact angles of aqueous dispersion with different contents of nanosilica particle were measured, and the specific results are shown in Figure 5.

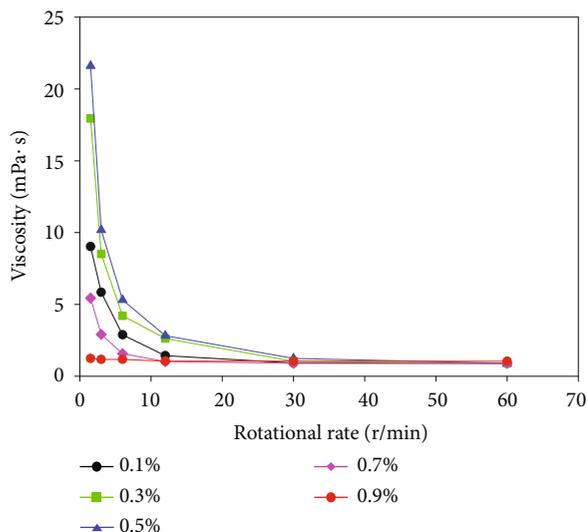


FIGURE 6: Shear thinning of nanosilica particle aqueous dispersion.

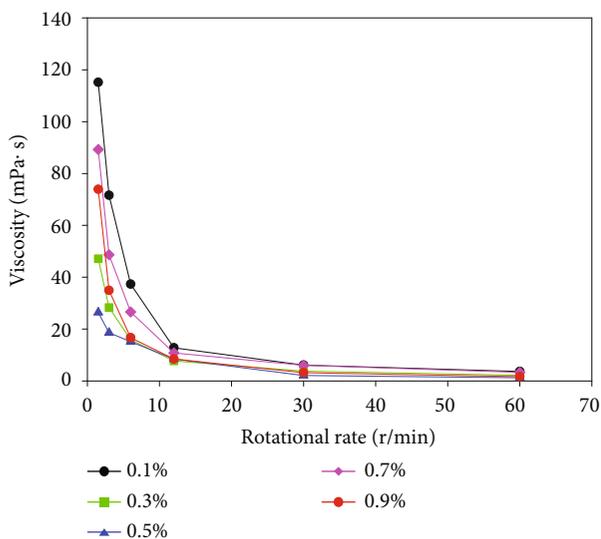


FIGURE 7: Relationship between the rotational rate and emulsion viscosity.

It could be seen from Figure 5 that with the increase in nanosilica particle content, the contact angle of aqueous dispersion with nanosilica particles showed a declining trend. When the content of nanosilica particles was 0.9%, the contact angle reached the minimum, about 25.56°. Since the measured contact angle is the angle between nanosilica aqueous dispersion and air on the interface of the glass slide and the main components of nanosilica particles and glass slide are basically the same, the adsorption amount of nanosilica particles on the surface of the glass slide increased with the increase in their content, and their adsorption capacity on the interface of aqueous dispersion and air was limited. Therefore, the three-phase contact angle increased with the increase in nanosilica particle content.

Figure 6 shows the relationship between the rotational rate and the viscosity of nanosilica particle aqueous disper-

sion at temperature 30°C. And the curves of different colors represented different contents of nanosilica particles. The aqueous dispersion of nanosilica particle had the significant phenomenon of shear dilution, and the viscosity decreased rapidly with the increase in rotational speed. The rotational rate of 30 r/min was the critical rate; when the rotational rate was less than 30 r/min, the viscosity of different nanosilica aqueous dispersions decreased rapidly with the increase in the rotational rate. In the study, when the content of nanosilica particle was 0.5%, the viscosity of O/W emulsion was maximum, followed by that of 0.3%, 0.1%, 0.7%, and 0.9%. When the content of nanosilica particle was 0.9%, the viscosity of O/W emulsion remained basically constant, and the rotational rate had little effect on the viscosity. Then, when the rotational rate was greater than 30 r/min, the viscosity of O/W emulsion was basically stable and no longer varied with the rotational rate, and the viscosity difference among O/W emulsions with different contents of nanosilica particle was small. And the viscosity of nanosilica particle aqueous dispersions was different when the content of nanosilica particle changed.

3.2. Effect Evaluation of Nanosilica Particles on O/W Emulsion Properties. After the effect of nanosilica particles on the interface properties of O/W emulsion was obtained, the stability, viscosity, and salt resistance of O/W emulsion reinforced by nanosilica particles were also systematically evaluated. And all the O/W emulsions in the study were prepared with the volume ratio between oil and water being 3:7.

3.2.1. Effect of Nanosilica Particles on Emulsion Stability. The O/W emulsions with the content of nanosilica particle among 0.1%~0.9% were prepared, and the effect of nanosilica particle on the stability of O/W emulsion was investigated. As a solid medium, nanosilica particles could adsorb on the oil-water interface to stabilize O/W emulsion. The observation period was ten days. During the first two days, the O/W emulsions were observed every two hours, and on the third day to the tenth day, the O/W emulsions were observed every half day. And after ten days, the O/W emulsions reinforced by nanosilica particle were still evenly distributed and had good stability, without stratification or precipitation. The nanosilica particles adsorbed on the interface of O/W emulsion could form a network structure, and the composite structure could improve the stability of the interface film, thereby improving the stability of the emulsion. The stability of the emulsion increased with the increase in nanoparticle content.

3.2.2. Effect Evaluation of Nanosilica Particles on the Viscosity of O/W Emulsion. The relationship between the rotational rate and the viscosity of O/W emulsion reinforced by nanosilica particles is shown in Figure 7, and the viscosity was measured at temperature 30°C. The curves of different colors in Figure 7 represented the different contents of nanosilica particles. The viscosity of O/W emulsion decreased as the rotational rate increased, and when the rotational rate was greater than 30 r/min, the viscosity of O/W emulsion

with different contents of nanosilica particles was almost the same. However, when the rotational rate was less than 30 r/min, O/W emulsion with different contents of nanosilica particles showed the phenomenon of shear thinning. The viscosity of O/W emulsion was not positively correlated with the content of nanosilica particles, and it showed that the emulsion with the content of nanosilica particle 0.1% had the highest viscosity, followed by that with the content of nanosilica particle 0.7%, 0.9%, and 0.5%, and when the content of nanosilica particle was 0.3%, O/W emulsion had the lowest viscosity. The viscosity difference between emulsions was related to the content of nanosilica particle, its adsorption capacity on O/W interface, adhesion state, and emulsion particle size, and it was a complicated process.

3.2.3. Viscosity Comparison between Aqueous Dispersion and O/W Emulsion. The rotational rate of 12 r/min was adopted to measure the viscosity of the aqueous dispersion of nanosilica particles and O/W emulsion reinforced by nanosilica particles, and the content of nanosilica particles in both aqueous dispersion and O/W emulsion was 0.1%, 0.3%, 0.5%, 0.7%, and 0.9%, respectively. Besides, the viscosity of aqueous dispersion without nanosilica particles was taken as the reference. As shown in Figure 8, the viscosity of aqueous dispersion with nanosilica particles was a little higher than that without nanosilica particles, and with the content of nanosilica particles increasing, the viscosity of aqueous dispersion with nanosilica particles showed the trend of increasing firstly and then decreasing. And when the content of nanosilica particles was 0.5%, the viscosity of the aqueous dispersion reached the maximum, 2.82 mPa·s. When the content of nanosilica particles was 0.7%, the viscosity of the aqueous dispersion reached the minimum, 1.02 mPa·s, almost the same as that without nanosilica particles, and was about one-third of that with the content of nanosilica particles 0.5%. The viscosity difference between different aqueous dispersions was due to the uneven distribution of nanosilica particles in the dispersion.

The viscosity of O/W emulsion reinforced by nanosilica particles was obviously higher than that of aqueous dispersion while the content of nanosilica particles was the same. As for the O/W emulsion reinforced by nanosilica particles, when the content of nanosilica particles was 0.1%, the viscosity of O/W emulsion reached the maximum, 12.77 mPa·s. However, when the content of nanosilica particles was 0.3%, the viscosity of O/W emulsion reached the minimum, 7.71 mPa·s. When the content of nanosilica particles was higher than 0.3%, the viscosity showed the trend of increasing firstly and then decreasing. When the content of nanosilica particles was 0.1%, there were not enough nanosilica particles to stabilize O/W emulsion, so the system was unstable and the viscosity showed much higher. When the content of nanosilica particles increased, the O/W emulsion tended to be stable and the emulsion particles tended to be smaller, showing an increase in viscosity. And when the content of nanosilica particles was 0.9%, the nanosilica particles were excessive, and the equilibrium of the system was disturbed, showing a decrease in viscosity.

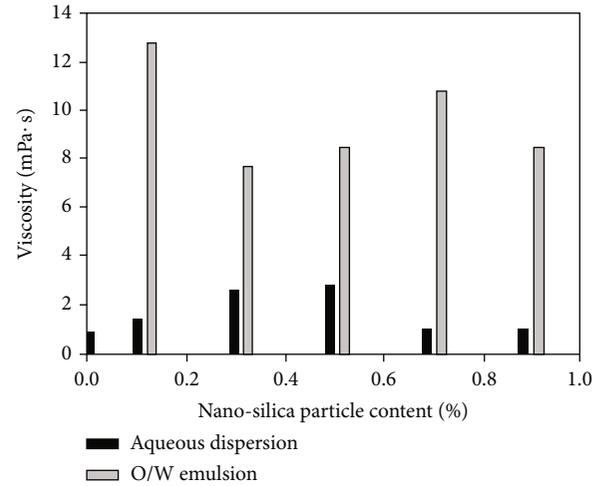


FIGURE 8: Viscosity comparison between aqueous dispersion and O/W emulsion.

3.2.4. Effect Evaluation of Salt Content on the Performance of O/W Emulsion Reinforced by Nanosilica Particles. From the above experimental evaluation, the stability and viscosity properties of O/W emulsion with the content of nanosilica particles 0.7% were better than those with other content, and it could significantly reduce both interface tension and contact angle. So during the evaluation, the content of nanosilica particles in O/W emulsion was chosen as 0.7%.

(1) Effect of Salt on Emulsion Stability. As for the effect of salt on the stability of emulsion, both magnesium salt and calcium salt were used to study their effects on the stability of emulsion, and the salt contents were selected as 0.008 mol/L, 0.010 mol/L, and 0.020 mol/L, respectively. The observation period was ten days, and during the first two days, the O/W emulsions were observed every two hours, and on the third day to the tenth day, the O/W emulsions were observed every half day.

After ten days of observation, the O/W emulsion was slightly transparent in the beaker with the magnesium salt content of 0.020 mol/L, followed by the magnesium salt content of 0.010 mol/L. Therefore, it could be found that the magnesium salt has the negative effect on the stability of O/W emulsion, and the higher the content of magnesium salt, the greater the effect on the stability of the emulsion. However, on the whole, the emulsion with different contents of magnesium salts still had good stability. With regard to the effect of calcium ion, it was found that with the increase in calcium ion, more transparent liquid was separated from the upper layer of O/W emulsion. By ten days, there were about 4 mm height precipitates in the beaker when the content of calcium ion was 0.020 mol/L, and there were obvious floccules in the stratification part. Therefore, calcium ion also had a certain inhibitory effect on the stability of O/W emulsion, and the effect was positively correlated with calcium ion content. On the whole, the effect of calcium salt on the stability of O/W emulsion was higher than that of magnesium salt.

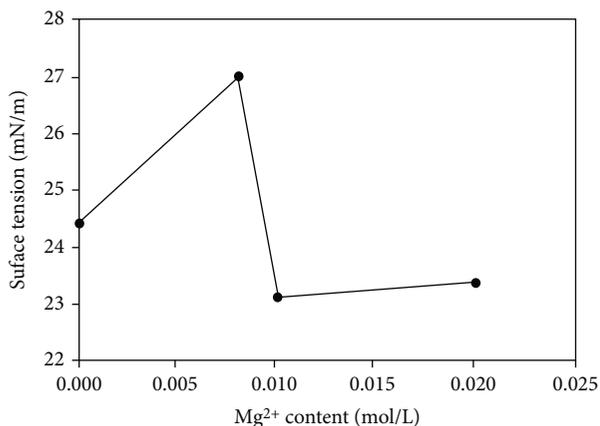


FIGURE 9: Relationship between Mg²⁺ content and emulsion surface tension.

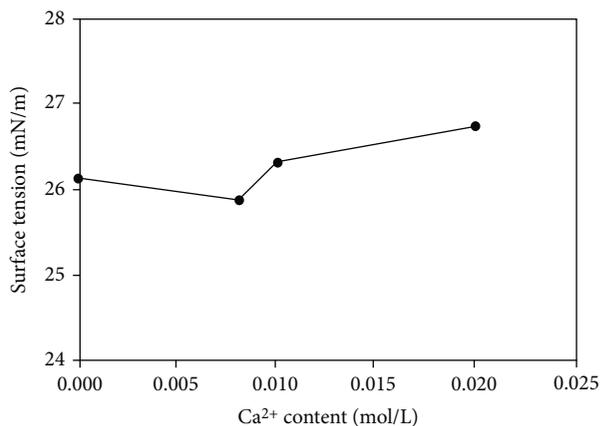


FIGURE 11: Relationship between Ca²⁺ content and emulsion surface tension.

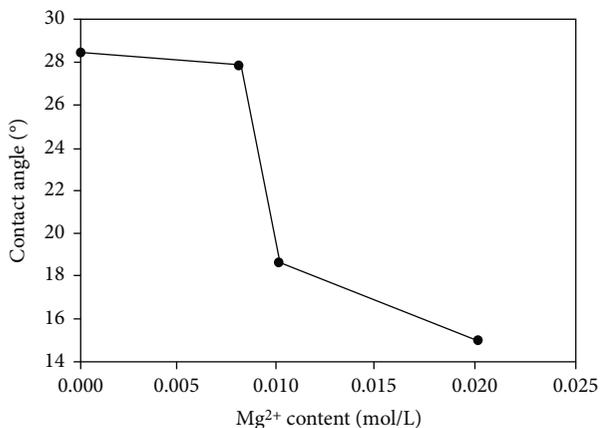


FIGURE 10: Relationship between Mg²⁺ content and emulsion contact angle.

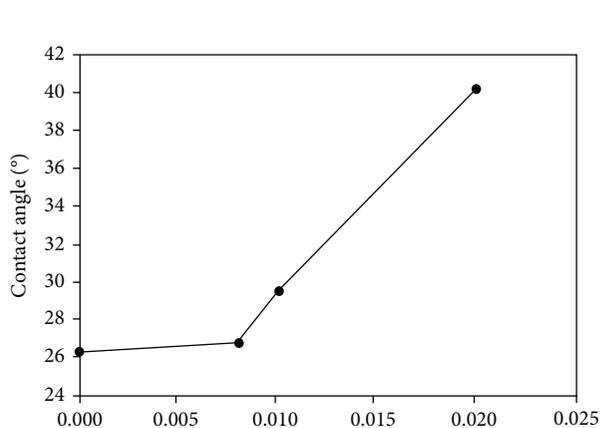


FIGURE 12: Relationship between Ca²⁺ content and emulsion contact angle.

The addition of inorganic salts was conducive to reducing the charge number of the interface film, compressing the thickness of the double layer, reducing the repulsion between the emulsion droplets, and facilitating the coalescence, thus effectively accelerating the demulsification of emulsions. The demulsification efficiency was related to the ionic valence state and the radius of hydrated ions.

(2) *Magnesium Ion (Mg²⁺)*. During the evaluation, the contents of Mg²⁺ were chosen as 0.008 mol/L, 0.010 mol/L, and 0.020 mol/L, respectively, and the effects of Mg²⁺ on surface tension, contact angle, and the stability of O/W emulsion reinforced by nanosilica particles were studied.

As shown in Figure 9, generally, with the increase in Mg²⁺ content, the surface tension of O/W emulsion reinforced by nanosilica particles first increased and then decreased and stabilized. When Mg²⁺ content was 0.008 mol/L, the viscosity of O/W emulsion reached the maximum, about 26.99mN/m. When Mg²⁺ content increased from 0.008 mol/L to 0.010 mol/L, the surface tension of O/W emulsion decreased greatly. After Mg²⁺ content was greater than 0.010 mol/L, the surface tension showed a stable trend.

As shown in Figure 10, with the increase in Mg²⁺ content, the contact angle of O/W emulsion reinforced by nanosilica particles generally showed a decreased trend, and Mg²⁺ content of 0.010 mol/L was taken as the critical point; when Mg²⁺ content was greater than 0.010 mol/L, the decreased amplitude was a little greater.

(3) *Calcium Ion (Ca²⁺)*. For the convenience of comparison, the content of Ca²⁺ in the experiment should correspond to the content of Mg²⁺, and the contents of Ca²⁺ were chosen as 0.008 mol/L, 0.010 mol/L, and 0.020 mol/L, respectively. The evaluation parameters were also the same, namely, surface tension, contact angle, and emulsion stability, respectively.

As shown in Figure 11, with the increase in Ca²⁺ content, the surface tension of O/W emulsion reinforced by nanosilica particles generally showed the trend of decreasing firstly and then increasing and being stable, which almost showed the opposite trend of the effect of Mg²⁺. The surface tension of O/W emulsion reinforced by nanosilica particles affected by Ca²⁺ was limited to 26 mN/m~27 mN/m, whose fluctuation was much smaller than that affected by

Mg²⁺. It showed that the surface tension of O/W emulsion was related to not only ion content but also ion type.

As shown in Figure 12, the contact angle of O/W emulsion reinforced by nanosilica particles generally increased with the content of Ca²⁺ increasing. When Ca²⁺ content was 0.020 mol/L, the contact angle was about 1.5 times that without Ca²⁺. Ca²⁺ content of 0.010 mol/L was taken as the critical point; when Ca²⁺ content was greater than 0.010 mol/L, the contact angle increased rapidly with the increase in Ca²⁺ content. However, while Ca²⁺ content was less than 0.010 mol/L, the contact angle increased slowly with the increase in Ca²⁺ content. So, as the content of Ca²⁺ increased, the hydrophobicity of O/W emulsion increased gradually.

4. Summary and Conclusions

- (1) The aqueous dispersion of nanosilica particles could significantly reduce the interface tension (with kerosene), and the extent of reduction was more than 50%. In terms of reducing the interface tension, the interface tension was 17.02 mN/m when the content of nanosilica particles was 0.9%. And the interface tension was 16.96 mN/m while the content was 0.1%. The decrease in interface tension was not positively correlated with the content of nanosilica particles
- (2) The aqueous dispersion of nanosilica particles could significantly change rock wettability. When the content of nanosilica particles increased from 0.1% to 0.7%, the contact angle decreased from 44.89° to 27.62°, showing a strong hydrophilic property. The relationship between contact angle and the content of nanosilica particles was not linear
- (3) The surface tension of O/W emulsion reinforced by nanosilica particles was among 25 mN/m~30 mN/m, which was at a low value. The contact angle was also very small, with the average of about 20.00° and a minimum of 12.50°, showing a strong hydrophilic property
- (4) The salts had little effect on interface tension of O/W emulsion reinforced by nanosilica particles but had a significant effect on the contact angle and stability of O/W emulsion. In the experiment, magnesium salt could reduce the three-phase (O/W emulsion, air, and glass slip) contact angle and increase the hydrophilic properties of O/W emulsion, while calcium salt had the opposite effect
- (5) Both calcium salt and magnesium salt could reduce the stability of O/W emulsion reinforced by nanosilica particles. The stability of O/W emulsion without either calcium salt or magnesium salt was about 10 days, but the oil-water stratification adding either calcium salt or magnesium salt was about 1 day~3 days earlier than that without salts; besides, calcium salt had a greater impact on the stability of O/W emulsion
- (6) When the nanosilica particle content was among 0.3%~0.7%, the viscosity of O/W emulsion increased with the increase in nanosilica particle content. When the content of nanosilica particles was 0.9%, the viscosity suddenly decreased, and the maximum extent of reduction was 21.7%

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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References

- [1] M. T. G. Janssen, R. M. Pilus, and P. L. J. Zitha, "A comparative study of gas flooding and foam-assisted chemical flooding in Bentheimer sandstones," *Transport in Porous Media*, vol. 131, no. 1, pp. 101–134, 2020.
- [2] S. Park, E. S. Lee, and W. Sulaiman, "Adsorption behaviors of surfactants for chemical flooding in enhanced oil recovery," *Journal of Industrial & Engineering Chemistry*, vol. 21, pp. 1239–1245, 2015.
- [3] J. E. Parra, G. A. Pope, M. Mejia, and M. T. Balhoff, "New approach for using surfactants to enhance oil recovery from naturally fractured oil-wet carbonate reservoirs," in *SPE Technical Conference & Exhibition*, Dubai, UAE, 2016.
- [4] Z. Wu, H. Liu, and X. Wang, "3D experimental investigation on enhanced oil recovery by flue gas coupled with steam in thick oil reservoirs," *Energy & Fuels*, vol. 32, no. 1, pp. 279–286, 2018.
- [5] K. Babu, N. Pal, V. K. Saxena, and A. Mandal, "Synthesis and characterization of a new polymeric surfactant for chemical enhanced oil recovery," *Korean Journal of Chemical Engineering*, vol. 33, no. 2, pp. 711–719, 2016.
- [6] H. Gao, H. Lai, J. Zhang, and Q. Xue, "Effects of bacterial cell density and alternating microbial- and enzymolysis-enhanced oil recovery on oil displacement efficiency," *Chemical Engineering Journal*, vol. 327, pp. 28–38, 2017.
- [7] L. Sierra, L. East, M. Y. Soliman, and D. Kulakofsky, "New completion methodology to improve oil recovery and minimize water intrusion in reservoirs subject to water injection," *SPE Journal*, vol. 16, no. 3, pp. 648–661, 2011.
- [8] D. Yin, Y. Q. Li, and D. F. Zhao, "Utilization of produced gas of CO₂ flooding to improve oil recovery," *Journal of the Energy Institute*, vol. 87, no. 4, pp. 289–296, 2014.
- [9] H. M. Zaid, N. R. A. Latiff, N. Yahya, H. Soleimani, and A. Shafie, "Application of electromagnetic waves and dielectric nanoparticles in enhanced oil recovery," *Journal of Nano Research*, vol. 26, pp. 135–142, 2013.
- [10] T. Guo, S. Zhang, Z. Qu, T. Zhou, Y. Xiao, and J. Gao, "Experimental study of hydraulic fracturing for shale by

- stimulated reservoir volume,” *Fuel*, vol. 128, no. 14, pp. 373–380, 2014.
- [11] N. Khan, J. Pu, C. Pu et al., “Comparison of acidizing and ultrasonic waves, and their synergetic effect for the mitigation of inorganic plugs,” *Energy & Fuels*, vol. 31, no. 10, pp. 11134–11145, 2017.
- [12] F. Medeiros, B. Kurtoglu, E. Ozkan, and H. Kazemi, “Analysis of production data from hydraulically fractured horizontal wells in shale reservoirs,” *SPE Reservoir Evaluation & Engineering*, vol. 13, no. 3, pp. 559–568, 2010.
- [13] M. Salah, M. El-Sebaee, and T. Batmaz, “Channel fracturing technology—a paradigm shift in stimulation of tight reservoir and unlock production potential,” in *79th EAGE Conference and Exhibition 2017- SPE EUROPEC*, Paris, France, 2017.
- [14] M. Wu, M. Ding, J. Yao, C. Li, Z. Huang, and S. Xu, “Production-performance analysis of composite shale-gas reservoirs by the boundary-element method,” *SPE Reservoir Evaluation & Engineering*, vol. 22, pp. 238–252, 2019.
- [15] B. Al-Shakry, B. S. Shiran, T. Skauge, and A. Skauge, “Enhanced oil recovery by polymer flooding: optimizing polymer injectivity,” in *SPE Kingdom of Saudi Arabia Annual Technical Symposium and Exhibition*, Dammam, Saudi Arabia, 2018.
- [16] M. S. Kamal, I. A. Hussein, and A. S. Sultan, “Review on surfactant flooding: phase behavior, retention, IFT, and field applications,” *Energy & Fuels*, vol. 31, no. 8, pp. 7701–7720, 2017.
- [17] I. I. Demikhova, N. V. Likhanova, A. E. Moctezuma, J. R. Hernandez Perez, O. Olivares-Xometl, and I. V. Lijanova, “Improved oil recovery potential by using emulsion flooding,” in *SPE Russian Oil & Gas Exploration & Production Technical Conference & Exhibition*, Moscow, Russia, 2014.
- [18] H. Zhong, T. Yang, H. Yin, J. Lu, K. Zhang, and C. Fu, “Role of alkali type in chemical loss and ASP-flooding enhanced oil recovery in sandstone formations,” *SPE Reservoir Evaluation & Engineering*, vol. 23, no. 2, pp. 431–445, 2020.
- [19] J. Wang, Y. Zhang, and J. Xie, “Influencing factors and application prospects of CO₂ flooding in heterogeneous glutenite reservoirs,” *Scientific Reports*, vol. 10, no. 1, 2020.
- [20] Y. Zhang, Y. Wang, F. Xue et al., “CO₂ foam flooding for improved oil recovery: reservoir simulation models and influencing factors,” *Journal of Petroleum Science & Engineering*, vol. 133, pp. 838–850, 2015.
- [21] B. Ding, Q. Sang, Z. Nie et al., “An improved study of emulsion flooding for conformance control in a heterogeneous 2D model with lean zones,” *SPE Journal*, vol. 26, no. 5, pp. 3094–3108, 2021.
- [22] X. Zhou, F. M. AlOtaibi, M. S. Kamal, and S. L. Kokal, “An experimental study on oil recovery performance using in situ supercritical CO₂ emulsion for carbonate reservoirs,” in *Abu Dhabi International Petroleum Exhibition & Conference*, Abu Dhabi, UAE, 2020.
- [23] C. C. Ezeuko, J. Wang, and I. D. Gates, “Investigation of emulsion flow in steam-assisted gravity drainage,” *SPE Journal*, vol. 18, no. 3, pp. 440–447, 2013.
- [24] M. Mojarad and D. Hassan, “Analytical modeling of emulsion flow at the edge of a steam chamber during a steam-assisted-gravity-drainage process,” *SPE Journal*, vol. 21, no. 2, pp. 353–363, 2016.
- [25] H. K. Sarma, B. B. Maini, and K. Jha, “Evaluation of emulsified solvent flooding for heavy oil recovery,” *Journal of Canadian Petroleum Technology*, vol. 37, no. 7, pp. 55–62, 1998.
- [26] J. Wang and M. Dong, “Simulation of O/W emulsion flow in alkaline/surfactant flood for heavy oil recovery,” *Journal of Canadian Petroleum Technology*, vol. 49, no. 6, pp. 46–52, 2009.
- [27] L. Cui and M. Bourrel, “Selection of CO₂-soluble surfactants for CO₂ foam/emulsion in hot and salty carbonate reservoirs,” in *RDPEURO 2018: Research and Development Petroleum Conference and Exhibition*, Dhahi, UAE, 2018.
- [28] A. Emadi, M. Sohrabi, S. A. Farzaneh, and S. Ireland, “Experimental investigation of liquid-CO₂ and CO₂-emulsion application for enhanced heavy oil recovery,” in *EAGE Annual Conference & Exhibition incorporating SPE Europec*, London, UK, 2013.
- [29] R. Hasannejad, P. Pourafshary, A. Vatani, and A. Sameni, “Application of silica nanofluid to control initiation of fines migration,” *Petroleum Exploration and Development*, vol. 44, no. 5, pp. 850–859, 2017.
- [30] Q. Sun, Z. Li, S. Li, L. Jiang, J. Wang, and P. Wang, “Utilization of surfactant-stabilized foam for enhanced oil recovery by adding nanoparticles,” *Energy & Fuels*, vol. 28, no. 4, pp. 2384–2394, 2014.
- [31] Y. Kazemzadeh, B. Dehdari, Z. Etemadan, M. Riazi, and M. Sharifi, “Experimental investigation into Fe₃O₄/SiO₂ nanoparticle performance and comparison with other nanofluids in enhanced oil recovery,” *Petroleum Science*, vol. 16, no. 3, pp. 112–124, 2019.
- [32] I. Kim, A. J. Worthen, M. Lotfollahi, K. P. Johnston, D. A. DiCarlo, and C. Huh, “Nanoparticle-stabilized emulsions for improved mobility control for adverse-mobility waterflooding,” in *SPE Improved Oil Recovery Conference*, Tulsa, Oklahoma, USA, 2017.
- [33] A. K. Alhuraishawy, R. S. Hamied, H. A. Hammood, and W. H. AL-Bazzaz, “Enhanced oil recovery for carbonate oil reservoir by using nano-surfactant: part II,” in *SPE Gas & Oil Technology Showcase and Conference*, Dubai, UAE, 2019.
- [34] H. A. Hammood, A. K. Alhuraishawy, R. S. Hamied, and W. H. AL-Bazzaz, “Enhanced oil recovery for carbonate oil reservoir by using nano-surfactant: part I,” in *SPE Gas & Oil Technology Showcase and Conference*, Dubai, UAE, 2019.
- [35] H. C. Lau, M. Yu, and Q. P. Nguyen, “Nanotechnology for oil-field applications: challenges and impact,” in *Abu Dhabi International Petroleum Exhibition & Conference*, Abu Dhabi, 2016.
- [36] O. A. Alomair, K. M. Matar, and Y. H. Alsaeed, “Experimental study of enhanced-heavy-oil recovery in Berea sandstone cores by use of nanofluids applications,” *SPE Reservoir Evaluation & Engineering*, vol. 18, no. 3, pp. 387–399, 2015.
- [37] G. Dordzie and M. Dejam, “Enhanced oil recovery from fractured carbonate reservoirs using nanoparticles with low salinity water and surfactant: a review on experimental and simulation studies,” *Advances in Colloid and Interface Science*, vol. 239, article 102449, 2021.
- [38] S. O. Olayiwola and M. Dejam, “A comprehensive review on interaction of nanoparticles with low salinity water and surfactant for enhanced oil recovery in sandstone and carbonate reservoirs,” *Fuel*, vol. 241, pp. 1045–1057, 2019.
- [39] O. Mahmoud, H. A. Nasr-El-Din, Z. Vryzas, and V. C. Keesidis, “Using ferric oxide and silica nanoparticles to develop modified calcium bentonite drilling fluids,” *SPE Drilling & Completion*, vol. 33, no. 1, pp. 12–26, 2018.
- [40] M. Rashidi, A. Sedaghat, B. Misbah, M. Sabati, and K. Vaidyan, “Use of SiO₂ nanoparticles in water-based drilling fluids for

- improved energy consumption and rheology: a laboratory study,” *SPE Journal*, vol. 26, no. 6, pp. 3529–3543, 2021.
- [41] R. Qalandari, A. Aghajanpour, and S. Khatibi, “A novel nanosilica-based solution for enhancing mechanical and rheological properties of oil well cement,” in *SPE Asia Pacific Oil and Gas Conference and Exhibition*, Brisbane, Australia, 2018.
- [42] A. K. Santra, P. Boul, and X. Pang, “Influence of nanomaterials in oilwell cement hydration and mechanical properties,” in *SPE International Oilfield Nanotechnology Conference and Exhibition*, Noordwijk, The Netherlands, 2012.
- [43] M. M. Tawfik, “Experimental studies of nanofluid thermal conductivity enhancement and applications: a review,” *Renewable & Sustainable Energy Reviews*, vol. 75, pp. 1239–1253, 2017.
- [44] A. W. Alsmail, A. Enotiadis, M. A. Hammami, and E. P. Giannelis, “Slow release of surfactant using silica nanosized capsules,” *SPE Journal*, vol. 25, no. 6, pp. 3472–3480, 2020.
- [45] C. Carpenter, “Gelled emulsions of CO₂, water, and nanoparticles,” *Journal of Petroleum Technology*, vol. 66, no. 7, pp. 135–137, 2014.
- [46] M. Khajehpour, R. Etminan, J. Goldman, F. Wassmuth, and S. Bryant, “Nanoparticles as foam stabilizer for steam-foam process,” *SPE Journal*, vol. 23, no. 6, pp. 2232–2242, 2018.
- [47] E. Neubauer, R. E. Hincapie, T. Clemens, and M. Cornelius, “Selection of nanomaterials as emulsion stabilizers in alkali-polymer EOR of high-TAN number oil,” in *SPE Improved Oil Recovery Conference*, 2020.
- [48] R. Singh and K. K. Mohanty, “Foams stabilized by in-situ surface activated nanoparticles in bulk and porous media,” *SPE Journal*, vol. 21, no. 1, pp. 121–130, 2016.
- [49] T. Uetani, J. Kai, T. Hitomi, H. Seino, K. Shimbori, and H. Yonebayashi, “Experimental investigation of crude-oil emulsion stability: effect of oil and brine compositions, asphaltene, wax, toluene insolubles, temperature, shear stress, and water cut,” *SPE Production & Operations*, vol. 35, pp. 320–334, 2020.
- [50] M. F. Fakoya and S. N. Shah, “Effect of silica nanoparticles on the rheological properties and filtration performance of surfactant-based and polymeric fracturing fluids and their blends,” *SPE Drilling & Completion*, vol. 33, no. 2, pp. 100–114, 2018.
- [51] W. Li, J. Liu, J. Zeng et al., “A critical review of the application of nanomaterials in frac fluids: the state of the art and challenges,” in *SPE Middle East Oil and Gas Show and Conference*, Manama, Bahrain, 2019.
- [52] L. Corredor, B. Maini, and M. Husein, “Improving polymer flooding by addition of surface modified nanoparticles,” in *SPE Asia Pacific Oil and Gas Conference and Exhibition*, Brisbane, Australia, 2018.
- [53] J. Theurer, O. Ajagbe, J. Osorio et al., “Removal of residual oil from produced water using magnetic nanoparticles,” *SPE Journal*, vol. 25, no. 5, pp. 2482–2495, 2020.
- [54] L. Hendraningrat, L. Shidong, and O. Torsæter, “A glass micromodel experimental study of hydrophilic nanoparticles retention for EOR project,” in *SPE Russian Oil and Gas Exploration and Production Technical Conference and Exhibition*, Moscow, Russia, 2012.
- [55] L. Hendraningrat, S. Li, and O. Torsæter, “Effect of some parameters influencing enhanced oil recovery process using silica nanoparticles: an experimental investigation,” in *SPE Reservoir Characterisation & Simulation Conference & Exhibition*, Abu Dhabi, UAE, 2013.
- [56] S. Li and O. Torsæter, “Experimental investigation of the influence of nanoparticles adsorption and transport on wettability alteration for oil wet Berea sandstone,” in *SPE Middle East Oil & Gas Show & Conference*, Manama, Bahrain, 2015.
- [57] M. Alaskar, M. Ames, S. Connor et al., “Nanoparticle and microparticle flow in porous and fractured media-an experimental study,” *SPE Journal*, vol. 17, no. 4, pp. 1160–1171, 2012.
- [58] A. Zamani, B. Maini, and P. P. Almas, “Propagation of nanocatalyst particles through Athabasca sands,” *Journal of Canadian Petroleum Technology*, vol. 52, no. 4, pp. 279–288, 2011.
- [59] T. Zhang, M. J. Murphy, H. Yu et al., “Investigation of nanoparticle adsorption during transport in porous media,” *SPE Journal*, vol. 20, no. 4, pp. 667–677, 2015.