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Experimental study and mechanism analysis of spontaneous imbibition of surfactants in tight oil sandstone

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Abstract:
The process of spontaneous imbibition is the basis of oil recovery from tight oil reservoirs. In this study, spontaneous imbibition experiments were conducted based on tight oil weakly hydrophilic sandstone cores from the Honghe oilfield in the Ordos Basin. Four different types of surfactants, such as nonionic Triton X-100, nonionic Tween-80, cationic dodecyl trimethyl ammonium bromide, and anionic sodium dodecyl benzene sulfonate, were separately dissolved in 30 g/L potassium chloride solution as simulated formation water. The effects of surfactant type on spontaneous imbibition were analyzed, and the results indicated that, because the nonions are adsorbed on the surface via Van der Waals force and adsorb H^+ through hydrogen bonds, the two nonionic surfactants altered the wettability of the core from weakly hydrophilic to strongly hydrophilic, the recovery rate was relatively high. The Triton X-100 was selected for subsequent spontaneous imbibition experiments by changing the mass concentration to adjust interfacial tension. It was found that the maximum recovery rate was 32% when the Triton X-100 mass concentration was 0.1%, which indicates that the enhanced recovery rate of spontaneous imbibition requires a sufficiently low wettability factor and a suitably high interfacial tension factor. Finally, the surfactants mixed with 0.03% sodium dodecylbenzene sulfonate and 0.1% Triton X-100 were used for spontaneous imbibition, attaining an oil recovery of up to 45%, which was 21.6% higher than that of single-surfactant imbibition. It was established that the synergistic mechanism depends on the wettability alteration of nonionic surfactant facilitating the spontaneous imbibition, while the anion accelerates oil removal from the core by continuously encasing oil droplets in the aqueous phase. This paper provides a theoretical basis for the imbibition development of weakly hydrophilic tight sandstone with high-salinity formation water.

1. Introduction

Unconventional tight oil reservoirs have attracted increasing attention because of their significant oil reserves (Cai et al., 2022). Although the technologies of horizontal wells and hydraulic fracturing have been successfully applied to develop unconventional oil resources, the oil recovery factor is commonly less than 10% (Wang et al., 2019; Gupta et al., 2020; Moosavi et al., 2020). As an important method of improving oil recovery in unconventional oil reservoirs, spontaneous imbibition (SI) behaviors have been widely studied (Cai et al., 2021; Wang et al., 2022). Oil recovery by SI is affected by many factors, such as rock wettability, pore structures, and fluid properties (Sheng., 2017; Wang et al., 2019; Hou and Sheng., 2022). The SI process is relatively slow in unconventional oil reservoirs because of the tiny pore throat sizes, while for oil-wet unconventional oil reservoirs,

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imbibition cannot occur spontaneously (Cheng et al., 2019; Zhang et al., 2022). Surfactants can accelerate the SI process by changing the rock wettability from oil-wet to intermediate-wet or water-wet, and the amount of surfactant required is only small (Wang et al., 2021). Therefore, SI using a surfactant is a promising way to improve unconventional oil recovery, while it is of great significance in studying the mechanisms of the SI of surfactant solution into oil-saturated nanopores in a tight matrix for unconventional oil reservoir development.

Recently, some advancements have been made to develop surfactants that can lower interfacial tension (IFT). Shen et al. (2016) investigated three types of surfactants for SI and revealed that low surface tension can lower the Jamin effect, thereby increasing the movable oil and enhancing oil recovery from the core. Liu et al. (2020) reported that spontaneous-emulsifying surfactants exhibiting ultra-low IFT can improve oil recovery from oil-wet cores, and the recovery performance surpassed that of regular surfactants by altering wettability. Alvarez et al. (2018) proposed that although the reduction in capillary force is beneficial for releasing the trapped oil, it is still difficult to drive the oil in the small capillary pores due to gravity; therefore, IFT should not reach an ultra-low value (e.g., 10^{-3} mN/m) to improve oil recovery in tight reservoirs. Li et al. (2018) developed a theoretical SI model based upon a modified Lucas-Washburn equation, in which the capillary force and flow resistance of the oil phase increase with the IFT, whereas the imbibition volume initially rises and then declines. In addition, the optimal IFT is 1.913-2.116 mN/m. However, a unified conclusion on the mechanisms of interfacial tension for surfactant SI is lacking.

Many researchers have agreed that the wettability alteration of surfactant-induced capillary-driven SI is the major mechanism affecting oil recovery in tight oil reservoirs. Wettability denotes the tendency of fluid diffusion to the solid phase in the presence of an immiscible fluid, reflecting the affinity of fluid on the solid surface (Diao et al., 2021). Unconventional reservoirs such as shale oil and tight oil cores mostly exhibit mixed wettability (Alvarez et al., 2016; Hu et al., 2022). Different types of surfactants (e.g., anionic, nonionic and cationic) have been compared in laboratory experiments in terms of wettability alteration to improve oil recovery. Souayeh et al. (2021) reported that a low-salinity nonionic surfactant was effective in altering the wettability of oil-wet calcite toward the water-wet state. Meanhile, low-salinity anionic surfactant systems were only able to reduce the oil-wetness of the oil-wet calcite. Hou et al. (2016) revealed that the cationic surfactant cetyltrimethylammonium bromide (CTAB) is more effective than the nonionic surfactant octyl phenol ethoxylate Triton X-100 (TX-100) and the anionic surfactant sodium laureth sulfate (POE (1)) in altering the wettability of oil-wet sandstone surfaces. Das et al. (2020) found that the cloud points of nonionic secondary alcohol ethoxylate surfactants are raised by the addition of anionic sulfonates, and mixed-surfactant systems were identified that modified the oil-wet surface to a water-wet surface with final contact angles as low as 70°. The authors also proposed the nonionic surfactant-induced dewetting of oil films, and a model involving "coating" and "sweeping" mechanisms was proposed to explain the mechanisms of surfactant action (Das et al., 2018). Nguyen et al. (2014) revealed that nonionic surfactants exhibited the highest efficiency, followed by anionic surfactants, and cationic surfactants were the least efficient in oil recovery from oil-wet Bakken shale cores. Several researchers reported that the cationic surfactant CTAB is better than the nonionic TX-100 and anionic surfactant POE (1) (Zhao et al., 2018; Ahmadi et al., 2020). Liu et al. (2019) established that anionic surfactants were more favorable in the recovery of shale oil than nonionic surfactants, as they have a better effect on wettability alteration. They also found that anionic surfactants perform better in terms of SI recovery due to their high efficiency in the wettability alteration of different clay surfaces in shale oil. Owing to the varying mineral compositions of different geological reservoirs, the abilities of different surfactants to change the wettability of the core and the relevant mechanisms are different, and the researches in this area are not comprehensive.

The properties of surfactants are greatly affected by the salt concentration. Previous studies have shown that ionic surfactants can enhance their surface activity in solutions with appropriate salt concentrations, that is, the oil-water IFT and critical micelle concentration (CMC) can be reduced (Santos et al., 2009). At the same time, tight oil reservoirs contain not only crude oil but also formation water with a certain degree of salinity. Therefore, the imbibing fluid can be considered a surfactant in a solution with a certain degree of salinity. When the salinity is extremely low, water sensitivity will occur in the core; however, when it is extremely high, salt sensitivity will result (Tian et al., 2016). When the concentration of inorganic salts is considerably high, hydrophilic groups combined with sufficient inorganic salt backions will aggregate micelles, eventually separating the solids and leading to phase separation (Renoncourt et al., 2007), and leading to the drastically reduced ability of surfactant to dissolve crude oil. Many experiments have been conducted on the SI of surfactants in a solution with ultra-low salinity (Nasralla et al., 2018; Teklu et al., 2018; Mohammadi et al., 2019; Song et al., 2020). They also demonstrated that low-salinity water imbibition can improve oil production from tight formations/shales due to wettability alteration, and the core becomes gradually more water-wet as the brine salinity decreases. Even though, few studies have focused on the SI of surfactants in formation water that has usually high salinity. Cai et al. (2020) investigated the influence of salinity and mineral components on SI, and the results revealed that high salinity can limit the expansion of wetting clay minerals (such as the illite/smectite mixed layer, illite) and decrease the maximum imbibed liquid volume. Therefore, the SI characteristics and mechanisms of different surfactants in high-salinity formation water still need to be studied.

In this paper, due to the unclear SI characteristics and mechanism of different surfactants in tight sandstone reservoirs with high salinity, SI experiments were conducted using four different types of surfactants in tight sandstone reservoirs with high salinity in the Zhenjing block of Ordos Basin. The oil-water IFT of different surfactants at different concentrations was first measured. Four different types of surfactants were dissolved in the simulated formation water of 30 g/L potassium chloride (KCl) solution, and then the SI experiments on tight sandstone cores in the Ordos reservoir were carried out. Under the same IFT, the influence of different surfactant salt solutions on the contact angle change was analyzed. The two non-ionic surfactants altered the core wettability from weakly hydrophilic to strongly hydrophilic. The recovery rate of non-ionic TX-100 was higher than that of Tween-80, and the TX-100 system was selected as the best candidate for wettability alteration. By changing the surfactant concentration to obtain the different IFT, the influence of IFT on SI was analyzed with a similar wettability change. Finally, the non-ionic TX-100 was mixed with anionic sodium dodecylbenzene sulfonate (SDBS) to evaluate the synergic imbibition effect of mixed surfactants. Then, the imbibition mechanisms of the three types of surfactant and the mixed surfactant were summarized to provide a theoretical basis for the subsequent development of unconventional reservoirs such as tight oil.

2. Experiments

2.1 Materials

In the experiment, the tight core of well 153 in the Zhenjing Block of Ordos Basin was selected. The well depth was 2,342-2,356 m. The core lithology here is gray oil-spotted fine sandstone, with permeability ranging from 0.2-0.7 mD and porosity varying between 7% and 16%. The oil in the formation is a low-sulfur, light-weight conventional crude oil. The imbibition liquid is the surfactant added into the simulated formation water, which is 30 g/L KCl solution. Four types of surfactants were selected: one anionic surfactant, namely SDBS (Tianjin Windboat Chemical Reagents Technology Co., Ltd., pure for analysis), two nonionic surfactants, namely Tween-80 (Tianjin Windboat Chemical Reagents Technology Co., Ltd., chemically pure), and TX-100 (Shanghai Macklin Biochemical Technology Co., Ltd., chemically pure), and one cationic surfactant, namely Dodecyl Trimethyl Ammonium Bromide (DTAB) (Shanghai McLeanMacklin Biochemical Technology Co., Ltd, 99% purity). The simulated oil was kerosene with a density of 0.81 g/mL.

The two-phase IFT was measured through an automatic interface tensiometer (K100, Krüss, Germany). CMC was determined using the platinum plate method, and the contact angle was calculated through the hanging drop method. The imbibition recovery rate was measured via the volumetric method based on a self-made imbibition bottle.

2.2 Method and procedure

(1) The core samples were cleaned in a Soxhlet extractor using toluene, methanol and deionized water. Toluene and methanol required approximately 24 days to remove non-polar and polar components from the porous medium.

(2) Core drying: The cores were oven-dried at 60 °C for two days until a constant weight of the core plugs was achieved.

(3) Core saturation: The core was vacuumed for 8 h and saturated with kerosene. Subsequently, the sample was soaked in saturated liquid at a pressure of 10 MPa for more than 40 h according to the standard SY/ T5336-2006 core analysis method. Afterwards, the floating oil on the core surface was wiped, and the core mass was weighed. The saturated oil mass was obtained by subtracting the core mass before saturation from the saturated core mass, and then the initial core contact angle was measured.

(4) SI: The core was placed in a customized imbibition bottle with an oil collecting pipe in the upper of the bottle at room temperature (25 °C), and the appropriate surfactant salt solution was selected for the static imbibition test. The state of the core surface and the change in the oil column height in the imbibition bottle were recorded at equal intervals to calculate the oil recovery.

(5) Finally, the core was removed, the surface liquid was wiped, and the core contact angle after imbibition was measured. The change characteristics of wettability were described based on the change in the contact angle before and after imbibition.

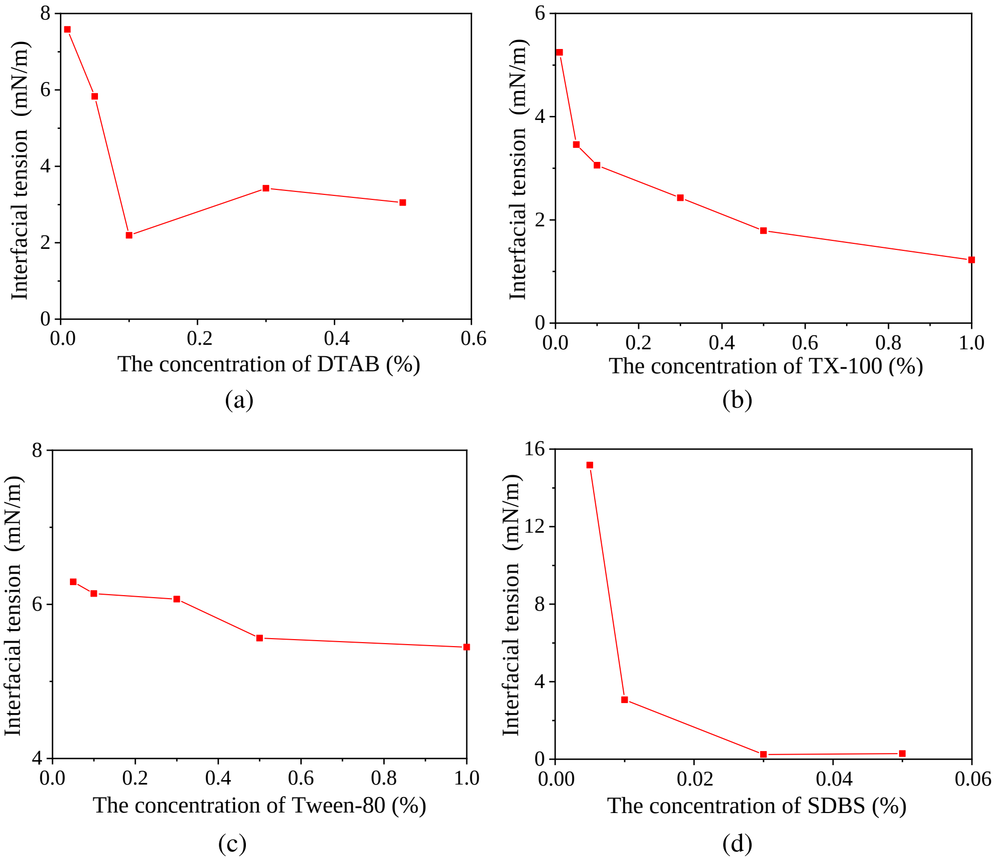
2.3 Oil-water IFT measurement

Four different surfactants were selected for this study: the cationic surfactant DTAB; the nonionic surfactants TX-100 and Tween-80; and the anionic surfactant SDBS. The four types of surfactant were prepared by dissolving in 30 g/L KCl salt solution, followed by the testing of their oil-water IFTs, as shown in Fig. 1. The ITF of 30 g/L KCl salt solution was measured as 24 mN/m, which was evidently lower than that of distilled water (72 mN/m).

Fig. 1(a) clearly shows the ITF of the DTAB salt solution in different mass concentrations (Note: all concentrations in this article refer to mass concentrations), with the increase in surfactant concentration from 0.01% to 0.1%, the IFT of oil-water first decreased from 6.5 to 2.1 mN/m; afterwards, ITF slightly increased to 3.5 mN/m at 0.3%, and then slightly decreased to 3.05 mN/m at 0.5%. The ITFs of the other three surfactants decreased with the increase in concentration. As the concentration of TX-100 increased from 0.01% to 1.0%, the oil-water IFT gradually decreased from 5.1 mN/m to 1.2 mN/m with a declining slope. As the concentration of Tween-80 increased from 0.01% to 1.0%, the oil-water IFT slightly lowered from 6.3 mN/m at 0.05% and reached the lowest value of 5.5 mN/m at 0.5%.

Fig. 1(d) shows the ITF of SDBS. As the SDBS had a low solubility in 30 g/L KCl solution, the surfactant crystal started to precipitate at a lower SDBS concentration. Therefore, four SDBS concentrations of 0.005, 0.01, 0.03, and 0.05% were selected, which were all below the phase separation concentration. The oil-water IFT for the SDBS salt solution decreased rapidly from 15 mN/m to 0.239 mN/m as the SDBS concentration increased from 0.005% to 0.03%, and remained unchanged until 0.05%. The concentration of SDBS surfactant dissolved in 30 g/L KCl solution was obviously limited, and the minimum IFT had a lower limit of 0.239 mN/m.

The above results revealed that all four surfactants could significantly decrease the oil-water IFT, and the SDBS in KCl solution had the lowest ITF, reaching below 1 mN/m. By measuring the IFT of these surfactants, an appropriate surfactant concentration could be selected for the SI experiments.



(a) (b)

(c) (d)

Fig. 1. Oil-water IFT between the different mass concentrations of surfactant salt solution and kerosene: (a) DTAB, (b) TX-100 (c) Tween-80, and (d) SDBS.

2.4 Core selection and contact angle measurements

Twelve cores were selected from the same well of HH153 in the Ordos tight oil reservoir, which had a cylindrical shape, diameter of 2.54 cm, and height of approximately 5 cm. The core parameters were measured and are listed in Table 1. The permeability (K) was determined via the gas permeability method, and the permeabilities of all the cores were between 0.23-0.68 mD, falling within the range of tight cores. The porosity was assessed via the high-pressure constant velocity mercury injection method, and ranged between 7% and 15%. All cores were selected after washing with oil, drying, and further saturating with oil. The saturation rates were all above 99%.

The contact angles of all cores were measured via the sessile drop method, with a schematic of the test shown in Fig. 2. Two contact angles were measured on the left and right surfaces of the core, and the average value was adopted. The contact angle before imbibition was measured by dropping deionized water on the saturated tight oil core surface after 1 min.

The experiments were divided into three groups: one group consisted of different surfactants with approximately the same ITF; the second group was set up for analyzing the influence of different ITFs; and the third group included two different surfactants for analyzing the combined effects. The core criteria for each group were identical permeability and porosity.

3. Results and discussion

3.1 Experiments for different surfactants

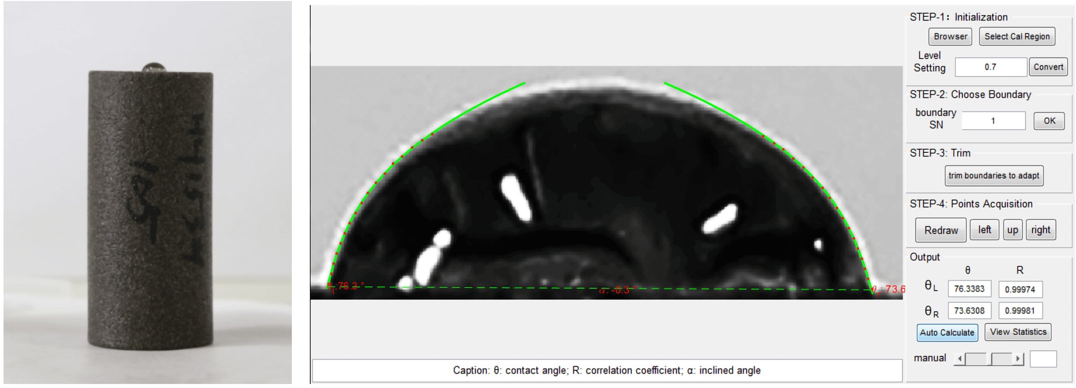
Five imbibition liquids were selected, including four surfactants separately dissolved in 30 g/L KCl solution and a blank contrast sample. The IFT of the samples was approximately 3 mN/m, except that the Tween solution had a value of 5.5 mN/m that was close to its minimum value of 5 mN/m, and the IFT of the blank KCl solution was 24 mN/m. The parameters of cores and imbibition liquids are listed in Table 1.

3.1.1 Experimental phenomenon

The imbibition experiments were conducted at room temperature (25 °C). All the cores were immersed in the imbibition liquid, and all sample surfaces were open for imbibition. To avoid the contact of the core bottom with the bottle bottom, the core was placed in a copper bracket. The data were recorded every 12 h, and photographs were obtained. The experiments were performed for a total of 220 h until the oil volume remained constant. A series of photographs of the imbibition process at different times for core HH153.58 in

Table 1. Core parameters in different groups.

No.	Core sample	Permeability (10^{-3} μm^2)	Porosity (%)	Saturation mass (g)	Saturation rate (%)	Initial contact angle (°)
I-1	HH153.105	0.626	10.6	2.2523	99.32	72.96
I-2	HH153.63	0.561	11.1	2.3922	99.59	55.31
I-3	HH153.45	0.653	15.0	3.1798	99.72	66.71
I-4	HH153.58	0.678	15.3	3.2835	99.76	69.11
I-5	HH153.104	0.618	10.5	2.2205	99.33	65.01
II-1	HH153.16	0.667	13.8	2.891	96.287	58.9
II-2	HH153.45	0.653	15.0	3.179	99.720	66.7
II-3	HH153.75	0.534	9.7	2.019	97.951	69
II-4	HH153.30	0.650	14.6	3.117	99.097	69.3
III-1	HH153.2	0.346	8.9	1.855	97.904	72.7
III-2	HH153. 5	0.306	8.7	1.641	99.293	65.8
III-3	HH153. 77	0.264	7.2	1.5415	99.395	75.0



STEP-1: Initialization

Level Setting: 0.7

STEP-2: Choose Boundary

boundary SN: 1

STEP-3: Trim

STEP-4: Points Acquisition

Output

	θ	R
θ_L	76.3383	0.99974
θ_R	73.6308	0.99981

Caption: θ: contact angle; R: correlation coefficient; α: inclined angle

Fig. 2. Tight core surface contact angle measurements.

0.5% Tween-80 KCl solution are shown in Fig. 3. Only after 12 h, part of the oil phase was clearly extracted depending on the imbibition process, and the oil phase was yellow. As time progressed, the height of the oil column increased, and the color of the oil phase gradually changed to light green. This color change was caused by the copper bracket and could be ignored. During the calibration of the volume of liquid column, the imbibition recovery represents the produced oil phase volume related to the saturated oil volume.

3.1.2 Analysis of results

Several factors such as the length, weight, surface area, and porosity of different samples affect the imbibition results. Owing to the difference in volume, porosity and density of the KCl solution, expressing the imbibition capacity simply using imbibed volume would be insufficient, and the raw imbibition data should be normalized. Thus, the dimensionless pore volume V_p was used to normalize the data for the imbibed liquid during imbibition from different samples. V_p represents the recovery rate R, which is expressed as follows:

$$V_p = R = \frac{HS\rho}{S_m} \tag{1}$$

where H denotes the height of the oil column entering the oil collecting pipe, S denotes the bottom area of the oil collecting pipe; ρ denotes the density of the imbibed liquid; and S_m denotes the saturated oil mass into the core. The relationship between the imbibition recovery rate of different imbibition systems over time is shown in Fig. 4.

As can be seen in Fig. 4(a), the 0.1% TX-100 system had the fastest imbibition speed and the highest imbibition recovery rate. The recovery of oil phase increased rapidly within the first 30 h, and practically stopped changing after 30 h, with a recovery rate reaching approximately 32%. The Tween-80 system had the second highest recovery rate of 25%; the imbibition rate was increased rapidly in the first 12 h, gradually increased from 12 to 144 h, and then remained stable. The recovery rate of the pure KCl solution system was ranked third. Although the recovery rate of the DTAB system was lower than that of the SDBS system from 24 to 160 h, it exceeded that of the SDBS system after 160 h. Therefore,

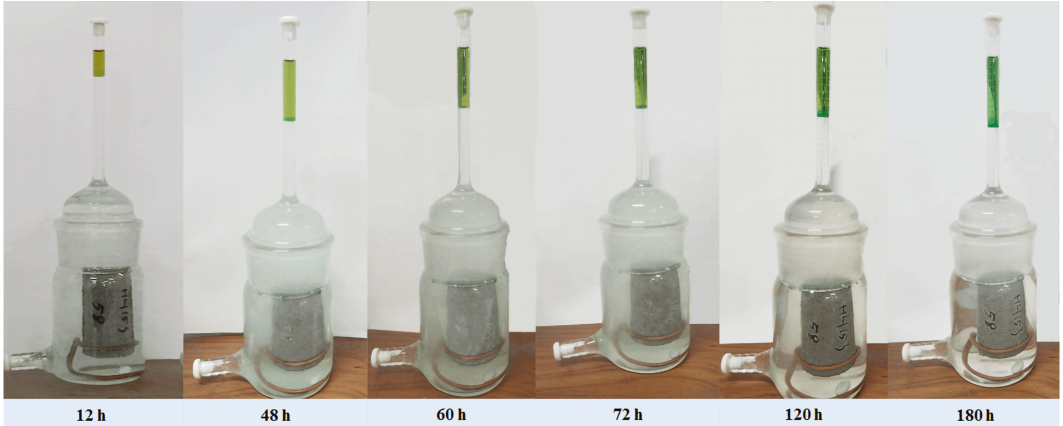
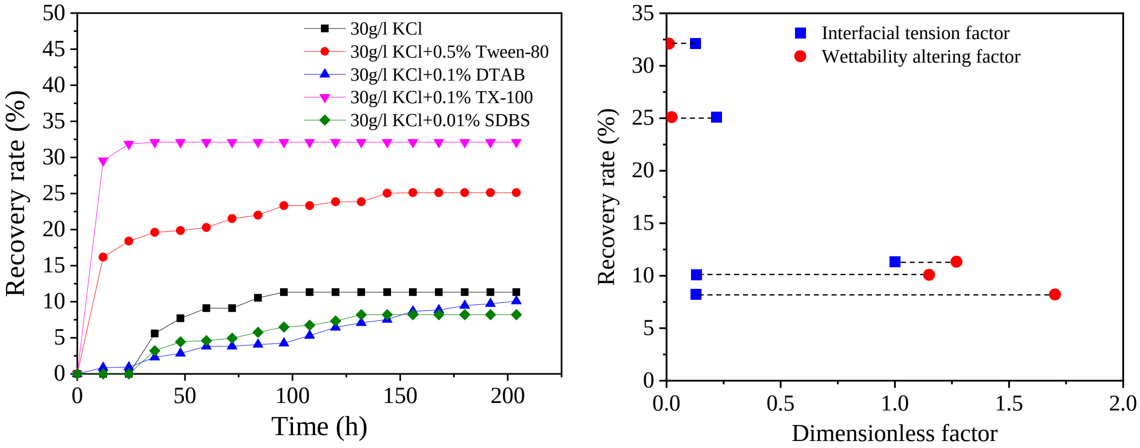


Fig. 3. HH153.58 core imbibition process over time.



(a) (b)

Fig. 4. (a) Relationship between the imbibition recovery rate of different imbibition systems over time and (b) Relationship between the ultimate recovery rates and different dimensionless factors.

the SDBS system exhibited the lowest ultimate recovery rate among all other surfactant systems.

Next, the change in the contact angle before and after imbibition was measured. The measurement steps of contact angle were as follows: the core was removed from the imbibition bottle and wiped to measure the contact angle of the deionized water droplets on the core surface. Then, changes in the contact angle of the core surface caused by the imbibing liquid were analyzed. The characteristic parameters such as the measured contact angle, IFT, and oil recovery rate are listed in Table 2.

Table 2 shows that the initial surface contact angle of the cores in this experiment was between 55° and 75°, indicating that all the cores were weakly hydrophilic or had intermediate wettability before imbibition. Moreover, the change in surface wettability was evidently different among the five imbibition liquid systems. Both the TX-100 solution and Tween-80 solution significantly changed the contact angle of the sandstone surface from weakly hydrophilic (60°-70°) to strongly hydrophilic ($< 10°$); the contact angle was reduced by approximately 60°. However, the contact angle of the core surface was increased by approximately 20° for the SDBS, 5° for the DTAB, and 10° for the pure KCl solution. The major driving force of spontaneous imbibition is capillary force F_c, which is expressed as follows:

$$F_c = \frac{2\sigma \cos\theta}{R_0} \tag{2}$$

where σ represents two-phase IFT, N/m; θ represents contact angle, °; R_0 is radius, m.

The relationship between the ultimate recovery rate and changed contact angle of the surfactant solution can be evaluated by the dimensionless factors as follows: IFT factor E_σ ($E_\sigma = \sigma_1/\sigma_0$), where σ_0 and σ_1 represent the IFTs at the pure KCl solution and under surfactant system, respectively. Herein, E_σ decreased with increasing ITFs of the surfactant system.

The wettability altering factor E_θ ($E_\theta = 1 - \cos\theta_1/1 - \cos\theta_0$), where θ_0 and θ_1 represent the contact angles before imbibition and after imbibition, respectively. Herein, E_θ decreased with decreasing contact angle in the surfactant system.

The values of these three factors for the five systems in this study are presented in Table 2 and Fig. 4(b). It can be seen that the wettability factor E_θ of the two nonionic surfactant

Table 2. Characteristic parameters and results for different imbibition systems.

No.	Imbibition fluid	IFT (mN/m)	Initial contact angle (°)	Final contact angle (°)	Interfacial tension factor	Wettability altering factor	Recovery rate (%)
I-1	30 g/L KCl	24	72.96	83.08	1	1.27	11.33
I-2	30 g/L KCl+ 0.01% SDBS	3.07	55.31	74.55	0.128	1.7014	8.21
I-3	30 g/L KCl+ 0.1% TX-100	3.06	66.71	6.59	0.1275	0.0109	32.11
I-4	30 g/L KCl+ 0.5% Tween-80	5.24	69.11	9.85	0.218	0.02285	25.1
I-5	30 g/L KCl+ 0.1% DTAB	3.14	65.01	70.39	0.131	1.15	10.08

systems is approximately equal to 0, and the recovery rate is higher than 25%; that is, the lower the wettability factor E_θ, the higher the recovery rate. The recovery rate of the TX-100 system is 32%, which is higher than that of the Tween-80 salt solution system (25%), indicating that TX-100 is a better-matched reservoir in the imbibition process. In contrast, when the wettability factor does not decrease ($E_\theta > 1$), although IFT for SDBS and DTAB surfactant systems decreases significantly ($E_\sigma < 0.131$), the oil recovery is still considerably lower than 15%.

3.1.3 Imbibition mechanism of different surfactants

Fig. 5 shows that the different surfactant systems exhibited different phenomena and recovery results. At 12 h of imbibition, both the pure KCl salt solution and the anionic surfactant SDBS salt solution systems had small oil droplets on the core surface, and no oil phase was present in the upper liquid column. As time progressed, although the size of oil droplets produced by imbibition gradually increased, most of them still were stuck to the core surface and were not easily detached, thereby decreasing the amount of oil entering the upper column of the imbibing bottle. For the pure KCl salt solution system, the IFT was high, and the droplets did not fall off easily from the surface because the oil phase and water phase are incompatible, implying that high IFT can retain the quality of the droplets. For the SDBS system, as the anionic surfactant was repelled by the negatively charged silicate core surface, it was difficult for anions to act on the core, and its SI characteristics were the same as that of a pure KCl system. The systems of nonionic surfactants, TX-100 and Tween-80 systems, produced small drops of oil that could easily detach from the core surface. The TX-100 system had a clean core surface with the greatest oil column height, and the oil droplets did not adhere to the core surface after imbibition. After 12 h, the core surface of the Tween-80 system was covered with numerous tiny oil droplets. Over time, the number of tiny oil droplets on the surface increased and they gradually formed an oil film. The height of the produced oil phase liquid column of the Tween-80 system was only second to that of the TX-100 system, which indicated that the nonionic surfactants were well adsorbed by the core and helpful for oil droplets to peel off from the core surface and enter the aqueous solution. For the DTAB surfactant salt solution, the oil droplets produced were small in size and number, and most of them were stuck to the core surface, which indicates that the DTAB was adsorbed by the sandstone core surface.

The core imbibition mechanisms of pure KCl solution, anionic SDBS, nonionic Tween 80 and TX-100, and cationic ion TDAB were revealed in Fig. 6. Fig. 6(a) demonstrates that the water in KCl solution enters the core partially through the action of capillary force, replacing part of the oil phase in the core. As the wall is weak hydrophilic, the oil phase is easily adsorbed on the wall. Due to the high IFT, the size of oil droplet on the wall is large and it is difficult to peel off the wall.

As shown in Fig. 6(b), the anionic surfactant is far away from the wall and difficult to adsorb on the core surface because of the electrostatic repulsion with negative ions on the core surface; hence, the result is similar to that of pure KCl system. Although anions have emulsifying properties and reduce IFT, they exhibit the lowest recovery rate.

As can be seen in Fig. 6(c), the head groups of nonionic surfactants carry no charge, which causes no electrostatic repulsion between the surfactant molecules and rock surfaces. Meanwhile, they can also adsorb on the surface of the core via van der Walls dispersion attraction, and the outward end binds to H^+ in water through hydrogen bonding so that the coated core surface becomes wet rapidly. The hydrophilic surface encourages water to enter the core and expel crude oil. Because the wall is hydrophilic, the discharged crude oil will not be adsorbed on the wall, and water will enter and collect at the top of the percolation bottle.

Fig. 6(d) shows that for the cationic surfactant DTAB, the cationic ion head is adsorbed on the surface of the negatively charged sandstone core, and the hydrophobic head is oriented towards the water phase, forming a typical oil-wet surface. Cations partially enter the interior of the cores, and the non-polar head of the surfactant binds to the oil phase in the core, forming the oil phase space, which cannot be discharged from the core; hence, the oil droplets are barely visible on the core surface and result in low recovery.

Overall, the nonionic surfactant system effectively changes the core surface wettability from weakly hydrophilic to strongly hydrophilic, and the other three solutions fail to

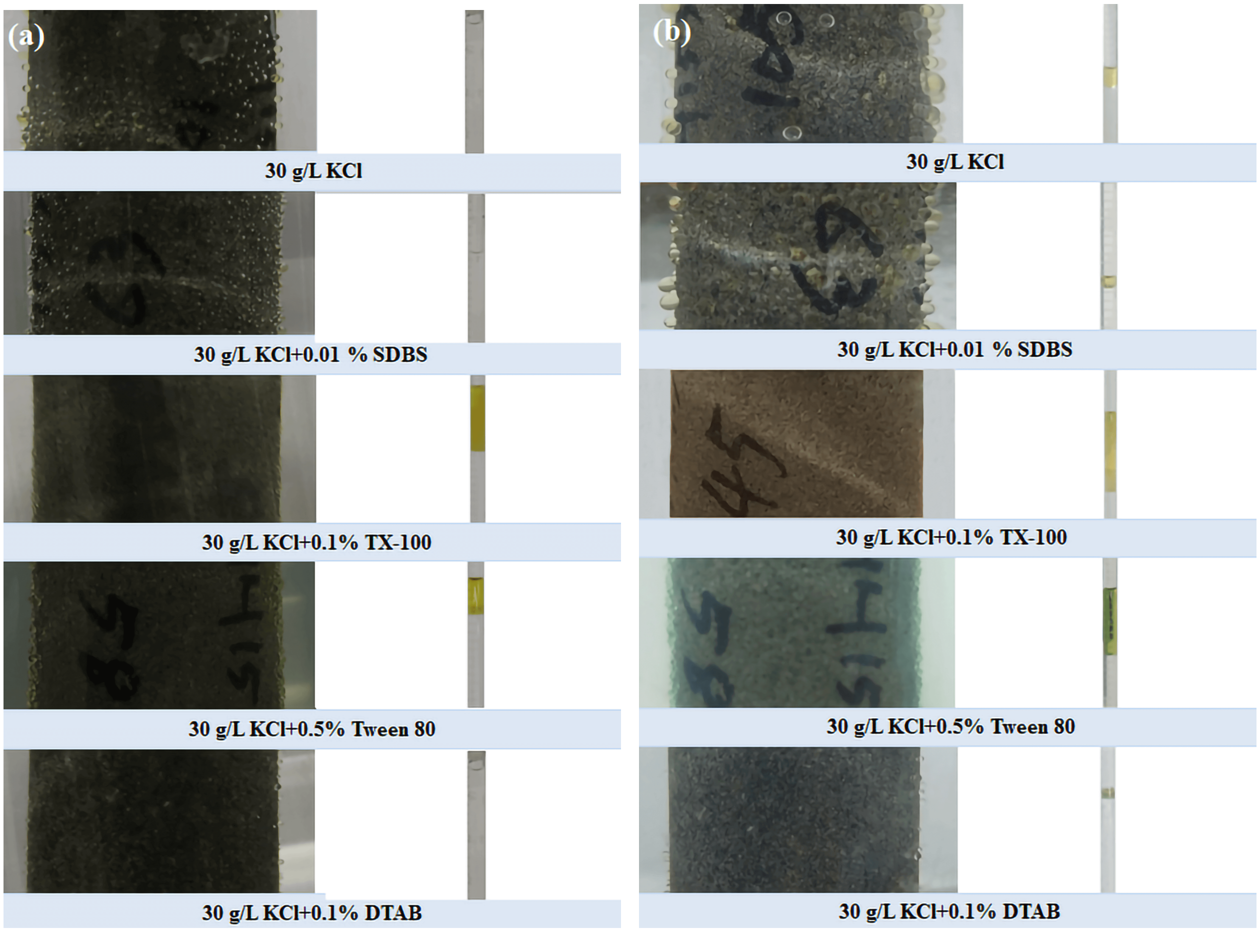


Fig. 5. Core surface morphology of the imbibition of different bottles and the liquid column height of produced oil at (a) 12 h and (b) 60 h.

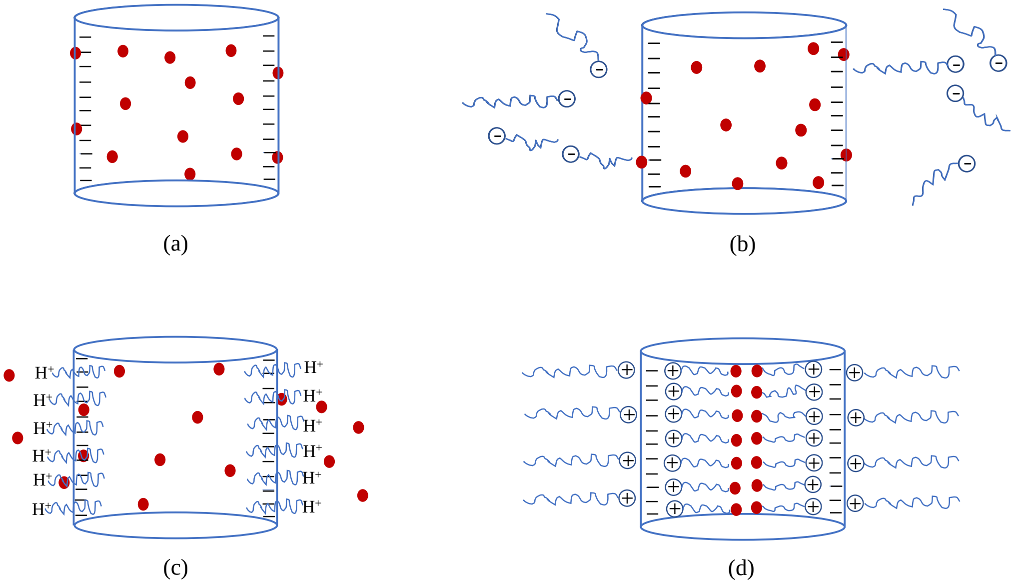
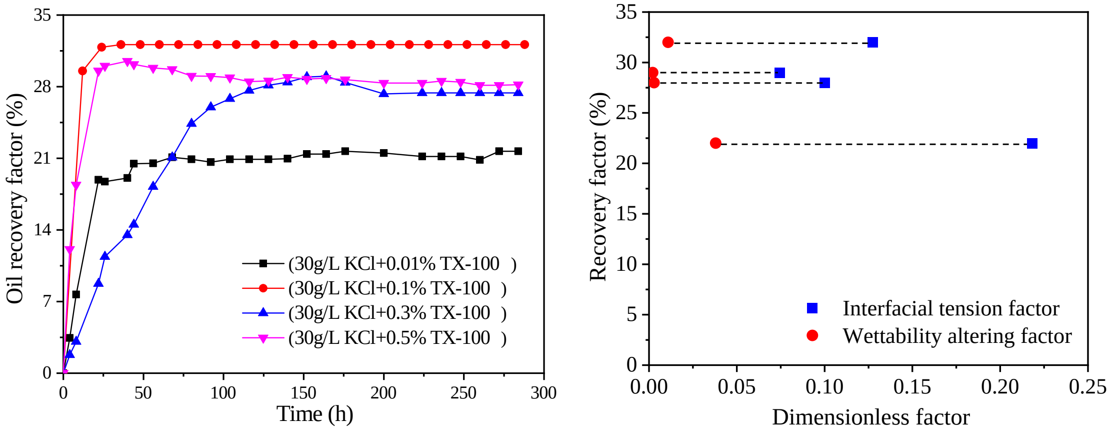


Fig. 6. Mechanism model of core imbibition with different kinds of surfactants: (a) pure 30 g/L KCl solution; (b) anionic surfactant, (c) nonionic surfactant and (d) cationic surfactant.



(a) (b)

Fig. 7. (a) Imbibition recovery processes corresponding to different TX-100 concentrations and (b) Relationship between the ultimate recovery rates and different dimensionless rates.

Table 3. Characteristic parameters and results for TX-100 imbibition system with different concentrations.

No.	Imbibition fluid	IFT (mN/m)	Initial contact angle (°)	Final contact angle (°)	Interfacial tension factor	Wettability altering factor	Recovery rate (%)
II-1	30 g/L KCL+ 0.01%TX-100	5.24	58.9	11	0.22	0.038	22
II-2	30 g/L KCl+ 0.1% TX-100	3.06	66.7	6.6	0.13	0.011	32
II-3	30 g/L KCl+ 0.3% TX-100	2.4	69	3.5	0.10	0.0029	28
II-4	30 g/L KCl+ 0.5% TX-100	1.79	69.3	3.0	0.075	0.0021	29

reverse the core wettability, thus the nonionic surfactant system achieves a higher recovery rate.

3.2 Effect of IFT for TX-100

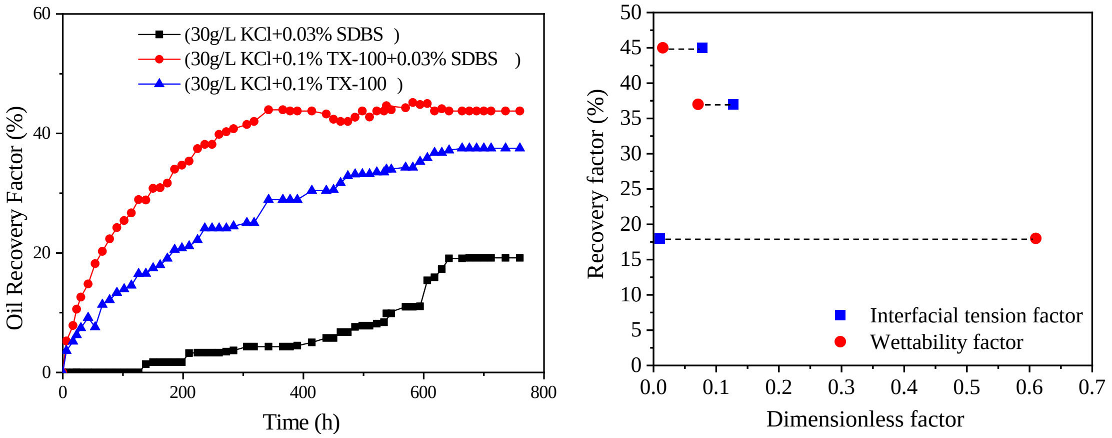
As determined by the experiment (Section 3.1), the imbibition recovery rate of the TX-100 system is higher than that of the Tween-80 system, and the oil-water IFT of the Tween-80 system negligibly changes with the concentration, whereas it significantly changes with the concentration for the TX-100 system. Therefore, imbibition experiments were conducted with different concentrations of TX-100 system to estimate the best concentration of TX-100 for imbibition. Cores II-1-II-4 were selected for this group of experiments, and the corresponding mass concentration of TX-100 was gradually increased from 0.01%, 0.1%, 0.3%, and 0.5%. The basic core data, such as permeability, are shown in Table 1. The permeability of all cores is approximately 0.6 mD; therefore, the influence of permeability on imbibition can be ignored.

As shown in Fig. 7(a), the recovery rate of the four systems in the test group increased rapidly at the initial stage, indicating a high recovery speed of imbibition oil. The imbibition of II-1, II-2, and II-4 was completed in the first 50 h, whereas II-3 required a longer time (approximately 150 h) to stabilize. The recovery rates of both II-3 and II-4 rapidly increased first and then slightly decreased. This is because the higher the concentration of TX-100, the stronger the emulsifying effect, and some water phases could be carried to the top of the suction bottle by microemulsion in the initial imbibition stage. As time progressed, the water phase gradually separated from the microemulsion phase, and the volume of the oil phase lessened.

As shown in Fig. 7 and Table 4, after imbibition, the order of recovery rate from high to low is II-2 (32%) > II-4 (29%) > II-3 (28%) > II-1 (22%). Obiously, the SI recovery rate of the 0.1% TX-100 system is the highest. This can be explained by the fact that for the 0.01% TX-100 system (II-1), the concentration is extremely low for altering the contact angle, and a higher value is not beneficial for imbibition. For II-2, II-3 and II-4, the values are all lower than 0.011, implying that the core becomes strongly hydrophilic as the concentration is higher than 0.1%, and the IFT decreases with the increase in concentration. However, a lower value is not beneficial for imbibition. The 0.1% TX-100 system (II-2) with the highest E_σ (0.13) under the condition of lower E_θ (0.011) achieves the highest recovery rate.

Table 4. Characteristic parameters and results for TX-100 imbibition system with different concentrations.

No.	Imbibition fluid	IFT (mN/m)	Initial contact angle (°)	Final contact angle (°)	Interfacial tension factor	Wettability altering factor	Recovery rate (%)
III-1	30 g/L KCL+ 0.03% SDBS	0.24	72.7	55.2	0.01	0.61	18
III-2	30 g/L KCl+ 0.1% TX-100+ 0.03% SDBS	1.88	65.8	7.6	0.078	0.015	45
III-3	30 g/L KCl+ 0.1% TX-100	3.06	75.0	18.7	0.13	0.071	37



(a) (b)

Fig. 8. (a) Recovery process of composite imbibition system and (b) Relationship between the ultimate recovery rates and different dimensionless rates.

3.3 Effect of mixed surfactants system

3.3.1 Experimental design

As described in Sections 3.2 and 3.3, the oil recovery rate of SI was not only affected by the oil-water IFT, but also closely related to the change in the contact angle of the core surface. As shown in Sections 2 and 3, the anionic surfactant SDBS can significantly reduce oil-water IFT, and the nonionic surfactant TX-100 can significantly reduce the core surface contact angle. A composite system was selected to examine the influence of mixed surfactants on SI. The selected core variables are presented in Table 1, while the experimental control groups are listed in Table 4.

3.3.2 Analysis of results

The change in the recovery process over time is shown in Fig. 8(a), and the relationship between the ultimate recovery rates and different dimensionless factors is presented in Fig. 8(b).

The influence of IFT and wettability on immobilization were evident from the experiment with this composite system. In this experiment, when the surface of the tight sandstone core was not sufficiently wet, that is, for the system of III-1 with E_θ of 0.61, the IFT was only 0.24 mN/m (E_σ (III-1) = 0.01), which could not effectively improve the imbibition recovery. For the 1% TX-100 imbibition system (III-3), the core was sufficiently wet, ($\theta = 18.7°$, $E_\theta = 0.078$), and the high IFT of 3.06 mN/m ($E_\sigma = 0.13$) could induce imbibition sufficiently with a high recovery rate 36%. However, for the composite system of III-2, the $E_\theta = 0.015$, core was extremely hydrophilic, the IFT had a moderate value of 1.88 mN/m ($E_\sigma = 0.078$), anthe imbibition recovery rate reached a maximum of 45%. This indicated that the low E_θ with moderate E_σ is the best condition for the enhancement of oil recovery.

In addition, the systems of III-2 (0.1% TX-100 + 0.03% SDBS) and II-4 (0.5% TX-100) were compared with the following results: the IFTs were 1.88 mN/m and 1.79 mN/m, the E_σ were 0.078 and 0.075, the E_θ were 0.015 and 0.0021, respectively. The E_σ values of the two systems were similar, and the E_θ were both lower than 0.015; however, the 45% recovery rate of III-2 was much higher than that of II-4 of 29%. The difference in recovery rate was large, indicating that if E_θ and E_σ are under similar conditions, it is necessary to compare the surfactant components in the system; that is, in addition to considering IFT and wettability, the interaction of multiple surfactants should also be considered.

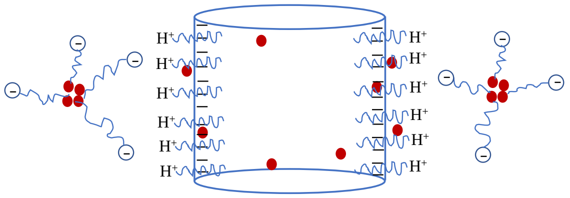


Fig. 9. Mechanism model of core imbibition with mixed surfactants, i.e., anionic surfactant and nonionic surfactant.

3.3.3 Imbibition mechanism of mixed surfactant systems

As can be seen from Fig. 9, after adding some anionic surfactants SDBS to the nonionic surfactant TX-100, as the core surface is altered into strong hydrophilic, water will displace the oil through capillary force, and the discharged oil will be stably adsorbed by the hydrophobic section of the anionic in the solution to form micelles. Thus, the dispersed oil phase will be reduced in the water phase, speeding up the oil phase expulsion from the core. This indicates the synergistic effect caused by the interaction of the two surfactants of TX-100 and SDBS. The synergistic mechanism lies in the wettability alteration of non-ions facilitating the SI, while the anion accelerates oil removal from the core by continuously encasing oil droplets in the aqueous phase.

4. Conclusions

In this study, SI experiments were conducted on tight oil sandstone cores from the Honghe oilfield in the Ordos Basin using four different types of surfactants dissolved in simulated formation water (30 g/L KCl solution). The influence of contact angle and IFT on SI oil recovery rate were analyzed by evaluating and , and the compound surfactants for the SI were investigated. The following conclusions were obtained from the experimental data:

The sandstone core surface was initially weakly hydrophilic. The recovery rates of the TX-100, Tween-80, DTAB and SDBS in salt solution systems were 32%, 25%, 10.08% and 8.21%, respectively. The nonionic surfactant TX-100 and Tween-80 can significantly reduce the surface contact angle of the core and greatly promote SI, that is, the lower the wettability factor (contact angle), the higher the recovery rate. The mechanisms of several surfactants are as follows. Cations enter the core and block oil droplets due to static electricity, while anions are far away from the wall, and are difficult to exert their effect. Nonions adsorb on the wall via Van der Waals force and adsorb H^+ through hydrogen bonds, thus changing the wettability of the wall and improving the imbibition recovery rate.

In the imbibition experiment with different concentrations of TX-100, the SI recovery was the highest at 32% for the 0.1% TX-100 system. It can be deduced that the wettability should be hydrophilic enough, and the IFT should not be very low, that is, sufficiently low (< 0.03) and high (> 0.07).

A composite system of 0.1% TX-100 + 0.03% SDBS was selected for SI. The contact angle was strongly hydrophilic ($E_\theta = 0.015$), IFT had a moderate value of 1.88 mN/m ($E_\sigma = 0.078$), and the imbibition recovery rate reached the maximum value of 45%, which was 21.6% higher than that for single-surfactant imbibition. The synergistic mechanism lies in the wettability alteration of non-ions facilitating the SI, while the anion accelerates oil removal from the core by continuously encasing oil droplets in the aqueous phase.

This paper provides new ideas for the development of tight oil in high salinity formation, especially nonionic surfactants that can provide good compatibility with the Honghe oilfield in the Ordos Basin: the nonionic TX-100 and anions SDBS, which have a good synergistic effect on SI. Our data provide guidance for the field development of tight oil reservoirs.

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Conflict of interest

The authors declare no competing interest.

Data Availability

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

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