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Research on Percolation Characteristics of a JANUS Nanoflooding Crude Oil System in Porous Media

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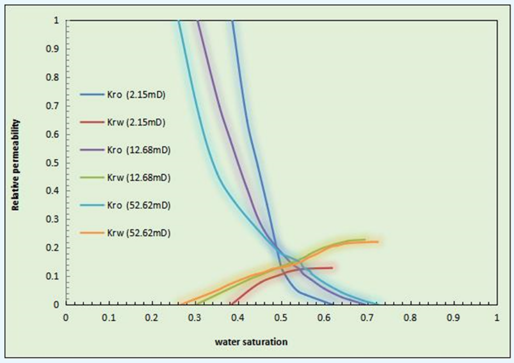
ABSTRACT: According to the research status of low-permeability reservoir development, in order to find an intelligent and efficient displacement method, a Janus smart nanocapsule (embedding nanomaterials with surfactant function into the polymer) is developed in this paper. There are two kinds of phase fluids in the migration of porous media, the initial Janus intelligent microcapsule slow-release liquid (dissolved and undissolved AP-g-PNIPAAM and Janus functional particle ternary dispersion) and the later AP-g-PNIPAAM and Janus functional particle binary dispersion. Comprehensively, using the indoor oil displacement experiment, the seepage characteristics of the Janus smart nanocapsule (JSNC) in porous media are studied, and their macro and micro oil displacement mechanisms are revealed. Research shows that Janus intelligent microcapsules have good mobility control ability in low-permeability heterogeneous reservoirs. The displacement performance of stepped differential pressure shows that the displacement medium can expand the swept volume. The research results presented can show that the JSNC oil displacement system has great application potential for the development of low-permeability reservoirs.



1. INTRODUCTION

Tight reservoirs have low permeability and porosity, and it is difficult for fluids to flow in porous media. Fracturing process and horizontal well development technology can effectively improve the conductivity of reservoir fluid and promote the growth of oil recovery,[1−3] but the increase is limited. With the entry of nanomaterials into the field of petroleum development, the size range of nanomaterials is internationally recognized as 1−100 nm.[4] In the petroleum industry, due to the compatibility of porous media with small-size materials, nanoscale and functionality have attracted the extensive attention of engineers.[5−7] Nano oil displacement agent has obtained good interfacial activity and oil displacement efficiency in some reports. However, the exploration of the percolation characteristics and oil displacement mechanism of nanomaterials is still in its infancy.

In a previous work, a Janus nanocapsule was developed. The capsule embeds Janus functional particles in the polymer to form a new binary composite system.[8,18] The capsule refers to the concept of medicine in medicine. The shell of the capsule can protect the components in the capsule from premature loss. In this paper, the shell of the capsule is a temperature responsive polymer, and the capsule is a nanosurfactant. This new binary composite structure can prevent the surfactant from being adsorbed near the well prematurely resulting in unnecessary waste. This is similar to the well-known medical capsule, so it is named nanocapsule.

The mechanism of Janus smart nanocapsule (JSNC) nanofluid changing the properties of crude oil and the synthesis scheme of JSNC are shown in Figure 1. The Pickering lotion method was used to modify nanomaterials. In previous studies, the nanomaterial used was silica, the oil phase emulsifier used was paraffin, and the nanomaterial modifier used was gamma-MPS. The response of JSNC nanoparticles obtained in the experiment was also confirmed in the structural characterization using infrared chromatography. The nanoparticles obtained by this method are convenient and fast, and the yield is considerable.

The modified nanoparticles can reduce reservoir pore water resistance and then effectively improve the injection volume of the displacement medium. The study of rheological properties of nanofluids is of great significance to the exploration of microdisplacement mechanism in low-permeability reservoirs. Hydrophilic-modified nanoparticles have the greatest reduction in capillary resistance, while hydrophobic-modified nanoparticles can form a superhydrophobic film on the rock surface. JSNC nanofluid is a kind of functional surfactant. Its

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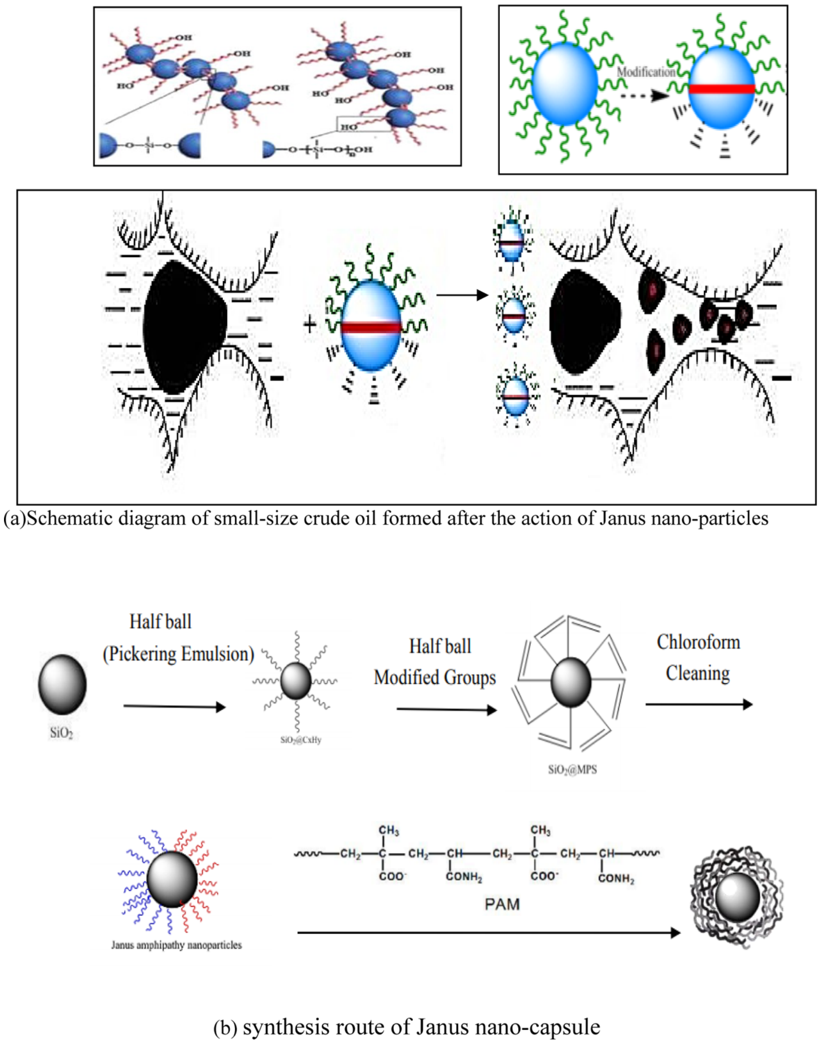

Figure 1. Synthetic route and action diagram of Janus nano oil displacement agent. (a) Schematic diagram of small-size crude oil formed after the action of Janus nanoparticles. (b) Synthesis route of the Janus nanocapsule.

unique properties are endowed by nanoscale and amphiphilic characteristics, which strengthen its ability to peel crude oil in porous media.[9−11] Amphiphilic nanoparticles will combine their own anisotropy and strong emulsification characteristics, give play to their own advantages, and irreversibly arrange at the oil−water interface. JSNC nanofluid, a surfactant-like material obtained by modifying nanoparticles, has strong emulsion stability. Based on its own structural characteristics, research shows that it is better than low-molecular-weight surfactants and uniformly modified nanoparticles.[12−16] Pickering emulsification allows small oil droplets to be bonded to the surface of solid particles.[7,17,18] The solid particles are closely connected with each other to avoid droplet polymerization and form a stable non chemical fusion emulsion. The Janus nano oil displacement system has strong interfacial activity and the ability to improve oil recovery of the tight reservoir.[19]

In order to further clarify the oil displacement mechanism of the Janus intelligent nanocapsule oil displacement system, the adsorption performance evaluation of JSNC, the comparative experiment of physical simulation of different injection systems, and the microvisual oil displacement experiment were carried out. The migration and oil displacement mechanism of JSNC in porous media are analyzed.

2. EXPERIMENT

2.1. Static Adsorption Experiment. *2.1.1. Experimental Conditions.* Experiment drug: 300 mL of Janus functional particle dispersion with mass concentrations of 0.05, 0.1, 0.15, and 0.3%, respectively; experimental sand: natural core oil sand (100−120 mesh); experimental temperature: 45 °C; equipment: water bath constant temperature oscillator (HD-SYC, Hunan Haode Instrument Equipment Co., Ltd., China); electric centrifuge (TG16B, Changzhou Jintan Liangyou Instrument Co., Ltd., China); temperature-controlled magnetic stirrer (85-2A, Shanghai Zhiwei Electric Appliance Co., Ltd., China); interface tensiometer (TX-500C, Kono Industry Co., Ltd.)

2.1.2. Experimental Method. Janus functional particle dispersion (0.05%) is taken as an example. 300 mL of Janus functional particle dispersion and 30 g (80−120 mesh) of oil sand were weighed and put into a 200 mL triangular flask. To this, 100 g of Janus functional particle dispersion was added. The triangular flask was sealed with a plug and adhesive tape, put in a shaking table, and shook evenly at 45 °C for 72 h. Then, it was taken and centrifuged at 4000 rpm for 20 min. The upper clear liquid was taken out. The absorbance of the centrifuged liquid was measured by an ultraviolet spectrophotometer, and the concentration was calculated by the standard curve formula, which is recorded as the concentration after primary adsorption, and the primary adsorption amount is calculated according to the formula. The above experimental steps were repeated to calculate the second and third adsorption amounts. The static adsorption capacity calculation formula is as follows

$$A_i = [(\rho_0 - \rho_1)V]/m \tag{1.1}$$

A_i—the nth adsorption amount, mg·g^{-1}; ρ_0—original concentration of the injected surfactant, mg·L^{-1}; ρ_1—surfactant concentration after the nth adsorption, mg·L^{-1}; V—rock particle quality, mL; m—rock particle quality, g.

$$A_n = \sum_{i=1}^{n} A_i \tag{1.2}$$

A_n—cumulative adsorption amount after the nth adsorption, mg·g^{-1}.

2.2. Dynamic Adsorption Experiment. *2.2.1. Experimental Conditions.* Experiment: Janus functional particle dispersion; experiment water: prepare simulated formation water (4500 mg/L); experiment: artificial core with permeability of about 15 × 10^{-3} μm^2; experimental temperature: 45 °C; experiment: pressure sensor; automatic thermostat, and so forth.

2.2.2. Experimental Method. The Janus functional particle dispersions with mass concentrations of 0.05, 0.1, 0.2, and 0.3% were prepared in 1000 mL each. The pore volume of saturated core was measured, and Janus functional particle dispersion with 10 times of pore volume was injected; Janus functional particle flooding was carried out, and the concentration of Janus functional particles at the outlet was detected.[20−24] The calculation formula of adsorption capacity is

$$Q = \left(C_0V_0 - \sum_{1}^{j} C_jV_j\right)/M \tag{1.3}$$

Q: adsorption capacity of Janus functional particles, mg·g^{-1}; C_0: implanted Janus functional particle concentration, mol·L^{-1}; V_0: the volume of Janus functional particles injected, mL; C_j: concentration of effluent from the j-th sampling bottle, mol·L^{-1}; V_j: the volume of effluent from the j-th sample receiving bottle, mL; M: the weight of core, g.

2.3. Seepage Characteristics of JSNC. *2.3.1. Experiment Condition.* Experimental core: artificial sandstone cylindrical core (2.5 cm × 2.5 cm × 10 cm); experimental temperature: 45 °C; experiment water: The salinity is 4500 mg/L of simulated water; experimental agent: JSNC, 0.2 wt %; experimental oil: simulated oil prepared by kerosene and dehydrated crude oil of an oil production plant in Daqing, room temperature viscosity 9.8 mPa·s.

2.3.2. Experimental Process. The core within the permeability range is selected, and the core pore volume is calculated. After recording, physical simulation indoor oil displacement experiment is carried out. The experimental temperature simulates the reservoir temperature of 45 °C, and the displacement speed is set to a constant speed of 0.1 mL/min to simulate the formation water to displace crude oil. The experimental data are recorded, and the relative permeability curve is drawn. The displacement experiments of 0.2 wt % Janus smart nanocapsule dispersion and 0.2 wt % ap-g-pnipaam polymer solution without Janus functional particles were carried out by using similar physical property cores. The above experimental process was repeated, and the relative permeability curve was drawn for comparative analysis.

2.4. Physical Simulation Experiments of Different Injection Systems. *2.4.1. Experimental Materials.* Experimental cores: artificial cores with water permeability of about 50 × 10^{-3} μm^2 are selected. Experimental oil: simulation oil (9.8 MPa s) prepared from degassed and dehydrated crude oil and kerosene in Daqing Oilfield; experiment water: distilled water, simulated formation water (4500 mg/L); experimental temperature: 45 °C; equipment: constant pressure and constant speed pump, core holder, piston container, vacuum pump, pressure gauge, and thermostat; experimental core: artificial core.

2.4.2. Experimental Scheme. Based on the sustained-release characteristics of Janus intelligent nanocapsules, the release time of capsules was shortened by adjusting the pH value of the system to acidity. Two groups of cores with basically the same parameters are set up to simulate the formation water flooding experiment; water flooding is carried out until the water cut is 98%, and the water flooding recovery ratio is calculated.

Laboratory oil displacement experiments were conducted in two groups to calculate the final recovery ratio. The relationship between the recovery rate increase and the system was studied.

The first group: 0.15 wt % Janus functional particles, 0.15 wt % Janus functional particles + 0.15 wt % AP-G-PNIPAAM, and 0.15 wt % Janus intelligent nanocapsules.

The second group: 0.15 wt % sodium alkylbenzene sulfonate and 0.15 wt % sodium alkylbenzene sulfonate + 0.15% AP-g-PNIPAAM.

2.5. Microvisual Oil Displacement Experiment. The microscopic visualization experiment can intuitively simulate the migration law of the system in porous media. In this paper, the laser lithography model is used to carry out the microscopic visualization experiment of Janus intelligent microcapsule dispersion system to study the migration of nanoparticles in porous media.

2.5.1. Experimental Materials. Experimental agent: Janus intelligent microcapsule dispersion system. Experimental oil: simulated oil prepared from degassed and dehydrated crude oil and kerosene in Daqing oilfield (9.8 MPa·s). Experimental water: distilled water, simulated formation water 4500 mg/L. Experimental equipment: microinjection pump, air compressor, szx16 microscope, ix73 differential interference difference biological microscope, image acquisition and processing system, lithographic glass model, measuring cylinder, pipeline, and so forth. Experimental temperature: constant temperature 45 °C.

2.5.2. Experimental Steps.

(1) The cylindrical small natural core with a diameter of 2.49 cm is selected, and the lithographic glass model is retained after oil washing and drying. The real pore structure of the copied small natural core is identified by laser technology, and then the pore structure is etched on a specific glass plate to make the lithographic glass model required for the experiment. The size of the model is 4.0 cm × 4.0 cm; the model has an injection end and a production end at the diagonal corner.

(2) After aseptic treatment, the lithographic glass model is dried and saturated with a micro syringe pump to simulate the formation. Water is then saturated to simulate oil, and a large number of bubbles are avoided in the process of saturation.
(3) Water drive stage: the simulated formation water is injected into the micromodel at a constant speed; the micro seepage process and residual oil distribution in the water drive stage in real time is observed, and the injection speed is 0.01 mL/h.
(4) Displacement stage of Janus smart microcapsule system: the Janus smart microcapsule is injected into the micromodel at a constant speed, the micro seepage process of the system is observed in real time, the injection speed is 0.01 mL/h, and the micromodel is observed regularly by a differential interference difference biological microscope.
(5) Subsequent water drive stage: the simulated formation water is injected into the micromodel at a constant speed, the microseepage process is observed in the subsequent water drive stage in real time, and the injection speed is 0.01 mL/h.

3. RESULTS AND DISCUSSION

3.1. Part of Static Adsorption Experiment. From the data in Table 1, it can be seen that with the increase of concentration, the single adsorption capacity and cumulative adsorption capacity both increase. The first adsorption amount is the highest, the second adsorption amount is lower, and the third adsorption amount is basically unchanged (Table 1).

Table 1. Static Adsorption Capacity at Different Concentrations

initial concentration (wt %)	concentration after primary adsorption (wt %)	concentration after secondary adsorption (wt %)	concentration after the third adsorption (wt %)
0.05	0.045	0.038	0.035
0.1	0.087	0.079	0.077
0.15	0.135	0.133	0.132
0.2	0.185	0.182	0.180
0.3	0.290	0.282	0.281

3.2. Part of Dynamic Adsorption Experiment. From the data in Table 2, the dynamic adsorption experiment shows that the dynamic adsorption capacity of Janus functional particle dispersion is much smaller than the static adsorption capacity. The main reason is that the specific surface area of static adsorption is larger than that of dynamic adsorption. At the same concentration, the initial adsorption capacity of Janus functional particle dispersion on the surface of rock particles increases rapidly during the injection process. When the accumulated adsorption amount reaches a certain value, the adsorption keeps equilibrium and the adsorption amount does not change. With the increase of injection concentration, the adsorption capacity of Janus functional particle dispersion on the surface of rock particles gradually increases.

When the injected mass concentration reaches a certain concentration, the adsorption capacity will not increase. The adsorption of rocks to Janus functional particle dispersion reached the upper limit. Static adsorption capacity and dynamic adsorption capacity show that the dynamic adsorption capacity is lower than the static adsorption capacity. This is because the adsorption of Janus functional particle dispersion is accompanied by desorption, and the contact area with the rock surface is small. In the dynamic adsorption process, with the continuous injection of the surfactant system, the adsorption equilibrium state will be reached. In conclusion, Janus functional particles have higher desorption energy.

3.3. Part of Seepage Characteristics of JSNC. Under the core conditions of different permeabilities, the remaining oil saturation of the core is different. The permeability is low, the remaining oil saturation is high, and the copermeability zone in the relative permeability curve has a narrow span. Based on the data of irreducible water saturation and isotonic point, it can be known that the wettability of core is water-wet which is shown in Figure 2.

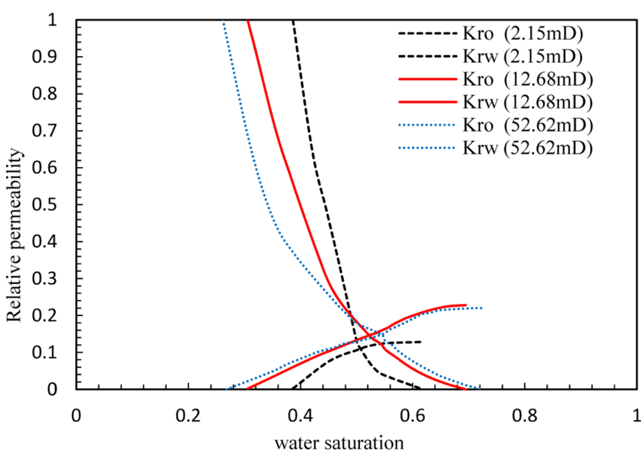

Figure 2. Janus smart nanocapsule dispersion system/oil relative permeability curve.

The dispersion system/oil phase permeability curve of Janus intelligent nanocapsules is observed. From Figure 2, in the permeability range of 2.25×10^{-3} μm^2 to 50.62×10^{-3} μm^2, the permeability increases, the irreducible water saturation and residual oil saturation of relative permeability curve decrease to varying degrees, the area of copermeability area increases, and the isotonic point moves to the left. It can be found from the obtained relative permeability curve. The relative permeability of oil phase is typical. Observing the position of the

Table 2. Dynamic Adsorption Capacity at Different Concentrations

core grade	original concentration (wt %)	diameter (cm)	length (cm)	porosity (%)	permeability 10^{-3} μm	post-adsorption concentration (wt %)
1	0.05	2.51	9.81	14.02	15.87	0.048
2	0.1	2.53	10.12	13.95	16.32	0.095
3	0.15	2.50	9.97	14.01	15.35	0.146
4	0.2	2.52	10.05	13.98	16.32	0.196
5	0.3	2.51	10.01	14.02	16.33	0.295

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isopermeability point, the curve trend of oil-phase relative permeability on the left side of the isopermeability point is steep, the trend shape of water-phase relative permeability curve on the right side of the isopermeability point is gentle, and the curve shape is very consistent with the curve characteristics of a low-permeability reservoir.

The experimental data show that the JSNC nanofluid makes the rising slope of water phase permeability slow, indicating that the nanofluid temporarily blocks the dominant channels in porous media, and more pore throat positions are affected in the flow. With the continuous migration of the JSNC nanofluid, the nanoparticles in the capsule in the JSNC fluid are released, resulting in a certain increase in the water-phase permeability, which is not very large on the whole.

As shown in Figures 3−5, the analysis shows that the water-phase permeability of the JSNC nanofluid decreases more than that of Janus nanomodified particle dispersion. Based on the interaction between Janus nanomodified particles and sandstone surface, the wettability of rock is reversed from hydrophilicity to lipophilicity, and the flow resistance of injected water is reduced, which greatly improves the permeability of injected water. Therefore, Janus nanomodified particles can play a role in increasing the injection amount in the flow of porous media. JSNC nanofluid can effectively reduce the permeability of aqueous phase. With the advancement of the displacement process, the nanocapsules will expand when flowing in porous media and release Janus nanomodified particles.

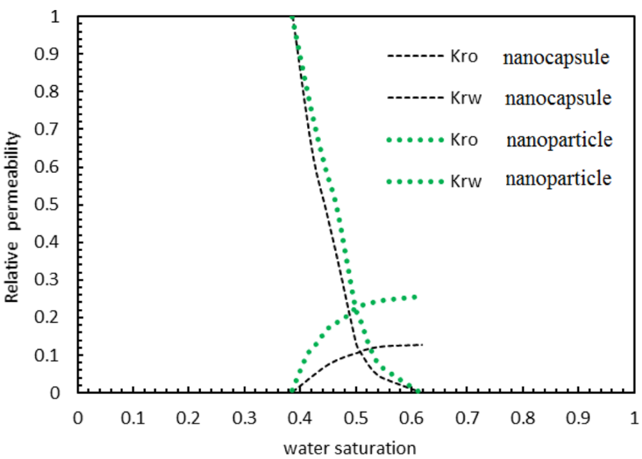

Figure 3. Comparison of Janus intelligent nanocapsule dispersion system (2.15 mD) and Janus functional particle (2.57 mD) oil relative permeability curve.

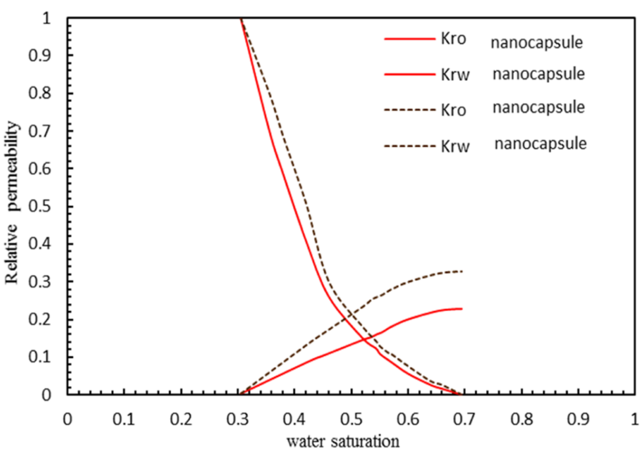

Figure 4. Comparison of Janus intelligent nanocapsule dispersion system (12.68 mD) and Janus functional particle (12.68 mD) oil relative permeability curve.

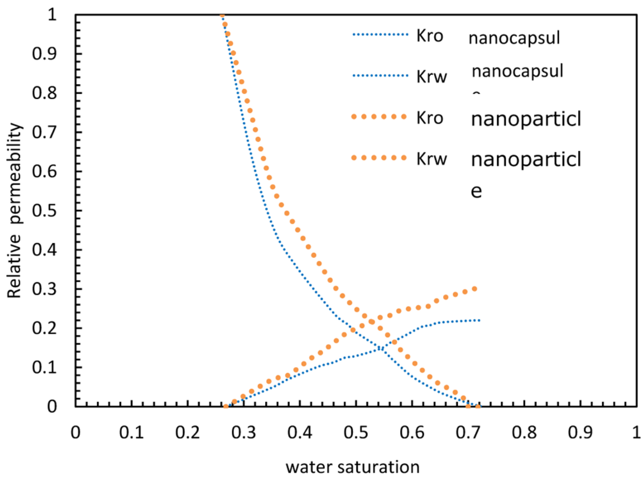

Figure 5. Comparison of Janus intelligent nanocapsule dispersion system (52.62 mD) and Janus functional particle (50.78 mD) oil relative permeability curve.

In JSNC nanofluid, the nanocapsule reaches the expansion limit under certain conditions. The expanded nanocapsule can increase the resistance, improve the water phase sweep area, reduce the water phase flow speed, and improve the oil displacement efficiency. When the nanocapsules stay in the pore throat of porous media, differential retention occurs, the direction of liquid flow migration changes, and the fluctuating pressure changes with fluid migration. In the path of JSNC propulsion, according to the laws of physics, the nanocapsules that temporarily block the larger pore throat release the modified nanoparticles at a constant speed. Through the relative permeability curve, we can find that the isopermeability point shifts, and the relative permeability and the isopermeability point of oil phase move to the right. This also confirms the above mechanism description. Relative permeability and isopermeability point of oil phase move to the right.

3.4. Part of Physical Simulation Experiments of Different Injection Systems. Experiments show that under the same experimental conditions, the oil displacement efficiency of capsule flooding is better than that of conventional binary combination flooding. 0.15 wt % Janus functional particle enhanced oil recovery is nearly twice as much as 0.15 wt % sodium alkylbenzene sulfonate after water flooding.

As shown in Table 3, on the basis of water flooding, Janus functional particles can improve oil recovery by 8.8%, and the final recovery ratio reaches 46.21%. After core injection, spontaneous emulsification occurs with the passage of time. It can be seen from the change of displacement pressure that the subsequent water injection resistance increases significantly. The results show that Janus functional particles can start the remaining oil in the core, expand the swept volume, and improve oil recovery. 0.15% Janus functional particles + 0.15% AP-g-PNIPAAM and 0.15 wt % Janus smart nanocapsules significantly enhance oil recovery, reaching 56.75 and 58.21%, respectively.

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Table 3. Displacement Data

oil displacement scheme	injection formula	recovery degree of water flooding (%)	EOR (%)	total oil displacement efficiency (%)
the first group	0.15% Janus functional particles	37.41	8.80	46.21
	0.15% Janus functional particles 0.15% AP-g-PNIPAAM	37.55	19.20	56.75
	0.15 wt % Janus intelligent nano capsule	37.67	21.54	58.21
the second group	0.15 wt % sodium alkylbenzene sulfonate	38.56	4.47	43.05
	0.15% sodium alkylbenzene sulfonate + 0.15% AP-g-PNIPAAM	37.69	14.42	52.11

3.5. Part of Microvisual Oil Displacement Experiment. The micrograph of saturated oil is shown in Figure 6. Figure 7 shows the distribution of remaining oil after PV water drive. As shown in Figure 8, the injected water has obvious water flow channels at the injection end and production end of the model. The channels are mainly concentrated at the diagonal between the injection end and the production end. The water flow migration process transits slightly to the other ends, but the area where the water flow does not affect is large.

Figure 6. Saturated oil glass plate.

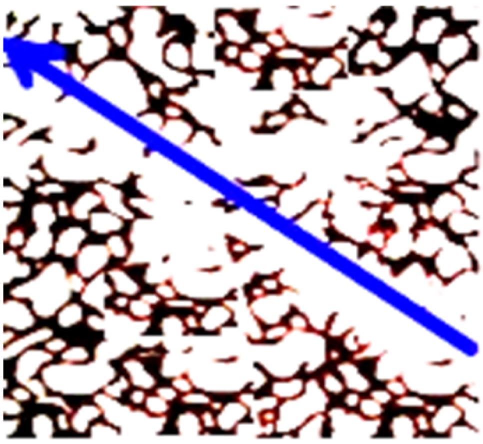

Figure 7. After 2PV water drive.

Figure 9 shows the distribution of remaining oil after Janus intelligent microcapsule was injected with 0.5PV. In the model, the water flows to the lower left and upper right regions. With the increase of injection volume, the liquid flow direction in the model changes many times, the model area is basically used, and the overall swept volume of the model is significantly improved. After the injected water forms a dominant channel, it starts ineffective water circulation displacement, the water flow sweep area is limited, and the remaining oil saturation in the reservoir is high. With the increase of the injection amount of Janus intelligent microcapsules in porous media, the injection system makes the model area basically used. Considering that the hydration expansion of the capsule has a certain role in regulating and blocking the large pores, the core material in the capsule is released with the increase of the injection amount, and the injection system forms Janus functional particle dispersion with polymer as the dispersion medium. The emulsification of Janus functional particles makes the injection system continue to contact with the residual oil during the migration process, and the emulsion droplets with a certain size and viscosity continuously migrate and accumulate. The specific profile control effect has been produced. The profile control effect of the emulsion greatly improves the sweep volume of the oil displacement system. At the same time, considering that the migration process of Janus

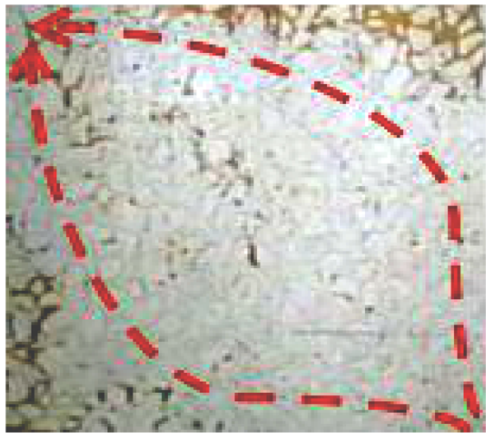

Figure 8. After displacement of Janus smart microcapsule.

Figure 9. After water drive.

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functional particles tends to drill into the hole throat, a large amount is filled and then the residual oil is squeezed out in the blind end, so as to turn the immovable oil into movable oil and improve the oil washing efficiency. Combined with the above factors, the reservoir residual oil is started, and the residual oil is transported to the outlet end by stripping and emulsification.

4. CONCLUSIONS

Through experimental verification, we can get the following knowledge, and the research results have reference significance for EOR research of heterogeneous tight reservoirs:

(1) The dynamic adsorption capacity of Janus functional particle dispersion is less than the static adsorption capacity. Considering adsorption and desorption, Janus functional particles have higher desorption energy.

(2) In the process of oil displacement, the competitive adsorption between Janus functional particles and polymer does not affect the interfacial activity of Janus functional particles, and the combination of Janus functional particles and polymer can effectively improve the oil displacement efficiency.

(3) After Janus nano oil displacement system, the relative permeability and isosmotic point of the oil phase shift to the right. The remaining oil saturation gradually decreases, the copermeability area widens, and the oil displacement efficiency is further improved.

(4) Janus intelligent nanocapsule dispersion system-oil relative permeability curve shows that the Janus intelligent nanocapsule dispersion system can effectively reduce water-phase permeability. When nanocapsules stay in the pore throat, the flow direction of the water phase changes. However, with the advancement of the displacement process, the nanocapsule temporarily locking the larger pore throat releases Janus functional particles as the core material in the capsule at a constant speed.

(5) Under the same experimental conditions, the oil displacement efficiency of capsule flooding is better than that of conventional binary combination flooding. After water flooding with 0.15 wt % Janus functional particles, the enhanced oil recovery is nearly 2 times higher than that of 0.15 wt % sodium alkylbenzene sulfonate.

(6) The results of the micro displacement experiment show that the Janus intelligent microcapsule system can improve the swept volume and enable the residual oil to start and carry out.

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Author Contributions

[§]F.S. and J.W. share first authorship. F.S. and J.W.: conceptualization and formal analysis. F.S.: investigation, methodology, validation, and writing—original draft preparation. J.W.: writing—review and editing. B.Z.: data curation and visualization.

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Notes

The authors declare no competing financial interest.

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