Fluoropolymer Microemulsion: Preparation and Application in Reservoir Wettability Reversal and Enhancing Oil Recovery

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ABSTRACT: Reservoir wettability is an important factor in the process of reservoir reconstruction. Especially in hydrophilic formation, it is easy to cause a water-locked phenomenon. A new type of fluoropolymer microemulsion was prepared by emulsion polymerization, and its structure and properties were characterized. The average particle size in the prepared emulsion was about 2.0 μm. The emulsion had good stability and wettability reversal performance for the storage of 30 days. After the treatment of 2.0 wt % emulsion, the contact angle between the core and water changed from 26 to 128°, the core surface free energy decreased from 66 to 2.6 mN/m, and the saturated water imbibition amount of the core decreased from 1.38 to 0.15 g. The ability of the fluoropolymer microemulsion to enhance oil recovery was evaluated by the visual displacement experiment. The fluoropolymer microemulsion can increase the displacement efficiency by more than 10%. The wettability of the core changed from hydrophilicity to hydrophobicity, and wettability reversal was achieved.
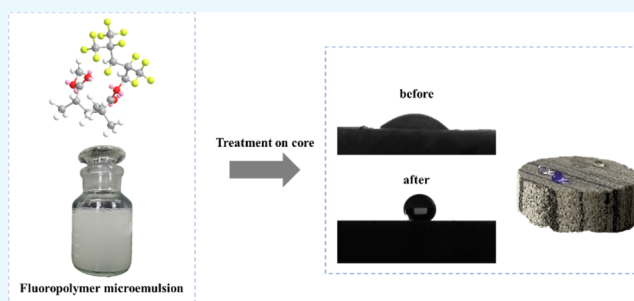
1. INTRODUCTION

The wettability of a reservoir plays an important role in oil and gas production and working fluid flowback.[1,2] In the process of oil and gas field development, a large amount of water-based working fluid is often needed.[3] In the reservoir with water wettability, the migration of the water phase in the reservoir has a great adhesion resistance. Furthermore, it causes a water-locked phenomenon in the reservoir, which increases the energy consumption in production and reduces the recovery of oil, gas, and working fluid.[4] In the process of drilling, welling, and production, the retention of the water phase in porous media is called a water-locked phenomenon or water lock effect. Water lock reduces the reservoir permeability and oil−gas relative permeability, which is not conducive to the exploitation of oil and gas resources. The strong water wettability of the reservoir is the main reason for this phenomenon.[5] Therefore, the transformation of reservoir wettability is one of the core problems in reservoir reconstruction. The change of the wettability of the reservoir from hydrophilicity to hydrophobicity can release the water-locked phenomenon.

The key problem to be solved in changing wettability is to change the free energy of a solid surface.[6] Generally speaking, the higher the surface free energy, the stronger the adsorption of the liquid on the surface, and vice versa.[7] The wettability of a surface can be changed in two ways. The direct way is to change the roughness of the surface. The wettability of the surface is enhanced by increasing the surface roughness.[8] For example, if the surface is hydrophobic, its hydrophobicity also

increases when the surface roughness increases.[9] Another way is to change the chemical structure of the surface material.[10] The wetting property of the reservoir can be changed by wetting inversion materials. Wetting reversal agents are mostly surfactant materials. This kind of reversal agent has good dispersibility.[11] But it lacks erosion resistance and adsorption capacity in the formation. Polymer materials can be used as wetting reversal agents to solve these problems.[12]

Fluoropolymers have very low surface free energy due to the small atomic radius of fluorine atoms and the large bond energy of the C−F bond in the structure.[13,14] Therefore, fluoropolymers are widely used in the preparation of interface materials with antiadhesion, antifriction, oil−water−gas separation, and etc.[15−17] But the preparation of fluorinated materials is also limited because of their extremely low surface free energy. The dispersity of fluoropolymers in water has hindered their application in the development of oil and gas fields.[18,19] Fluoropolymers can be prepared as emulsions to improve the dispersion in the water system.[20,21] As reversal agents, fluoropolymers are injected into the formation in the form of polymer fluids. After adsorption on the rock surface in
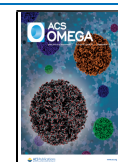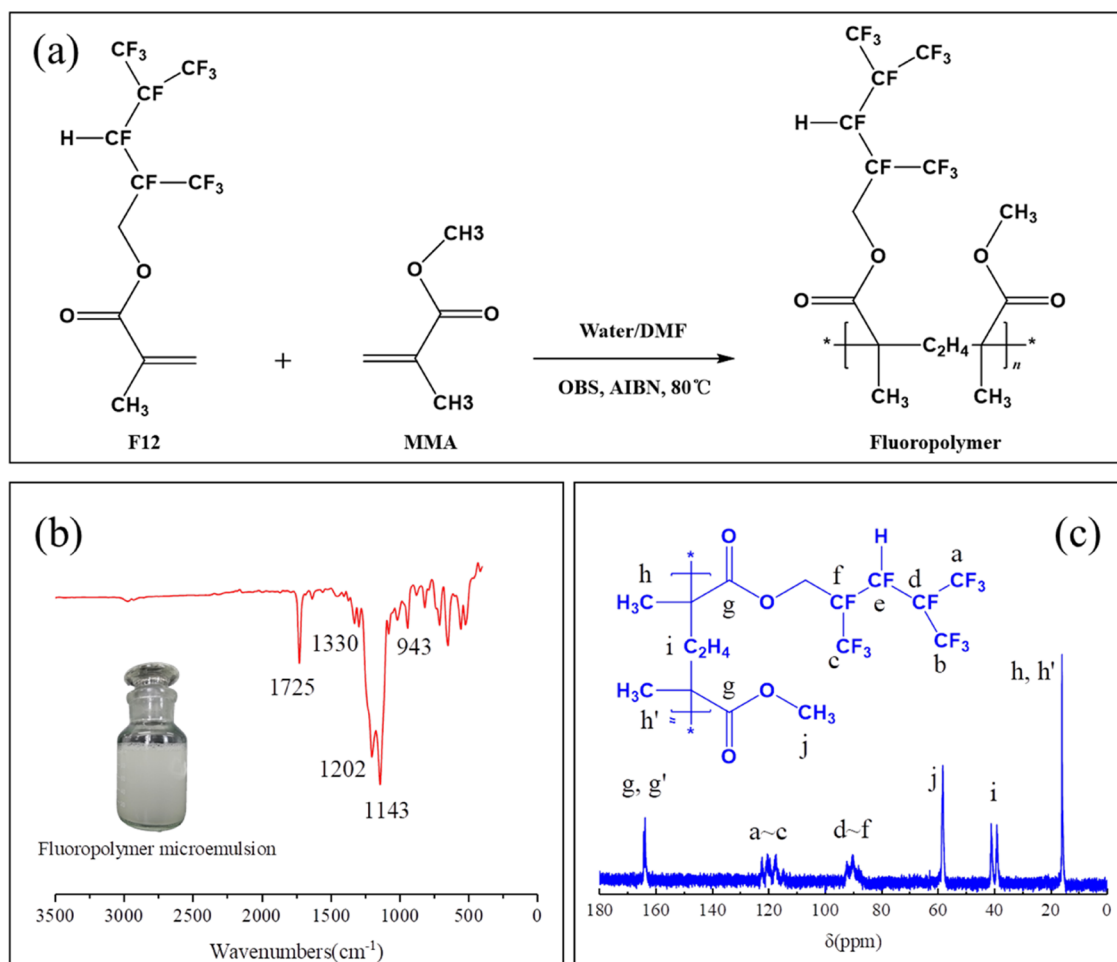
Figure 1. Reaction scheme of the fluoropolymer (a). Spectrogram of the fluoropolymer for (b) FT-IR and (c) ^{13}C NMR.

the formation channel, the wettability of the reservoir could be changed.

In our previous research,[22] we studied the wetting reversal of fluoropolymers with similar structures. In this work, a new type of fluoropolymer microemulsion was prepared by optimizing the polymerization system. The emulsion has micron-level particles and good dispersibility in the water system. The polymer emulsion can be used as a wettability reversal agent. We can change the wettability of the reservoir with low permeability and enhance the recovery efficiency and the recovery rate using the superhydrophobic property of the fluoropolymer. In the application of enhancing oil and gas recovery, it has reduced the surface free energy of the reservoir and changed the wettability from hydrophilicity to hydrophobicity.

2. RESULTS AND DISCUSSION

2.1. Characterization. The prepared fluoropolymer was separated and purified. The structure of the product was characterized by Fourier transform infrared (FT-IR) and ^{13}C NMR spectroscopies. Figure 1b shows the FT-IR spectrum of the product. The position of the peak at 1725 cm^{-1} belongs to the characteristic absorption peak of the C=O stretching vibration of the ester structure. The peaks at 1202 and 1143 cm^{-1} belong to the C−O stretching vibration in the structure. Wavenumbers of the peaks at 1330−943 cm^{-1} belong to the characteristic absorption peak of the C−F stretching vibration

of the fluoroalkane structure.[23] Figure 1c is the ^{13}C NMR spectrum of the product. The chemical shift at 170−160 ppm belongs to the characteristic peaks of ester carbon in the structure. The chemical shifts at 120 and 90 ppm belong to the characteristic peaks of fluorine-substituted carbon in the fluoropolymer.[24] Other peaks were also marked at the corresponding positions. Combined with the FT-IR and ^{13}C NMR results, the fluoropolymer is a block copolymer of F12 and MMA.

The size and shape of latex particles affect the performance of an emulsion.[25] The latex particles in the fluoropolymer emulsion can be roughly observed using a polarizing microscope. In the PM micrograph (Figure 2a), the bright spots are the polymer latex particles, and the dark spots are air bubbles in the emulsion. The latex particles distribute evenly in the emulsion and with size at the micron level. It can be observed more clearly using a transmission electron microscope (TEM). In the TEM micrograph (Figure 2b), the latex particles in the polymer emulsion are mostly spherical or ellipsoidal. Combined with the particle size curve (Figure 2c), the particle size distribution of the product is relatively concentrated, and the particle size is about 2 μm. The prepared product is a fluoropolymer microemulsion with uniform distribution of emulsion particles and strong stability.

The formation is usually at a high temperature and pressure. Organic chemicals cannot work because of the decomposition at a high temperature.[26,27] The thermal property of the product
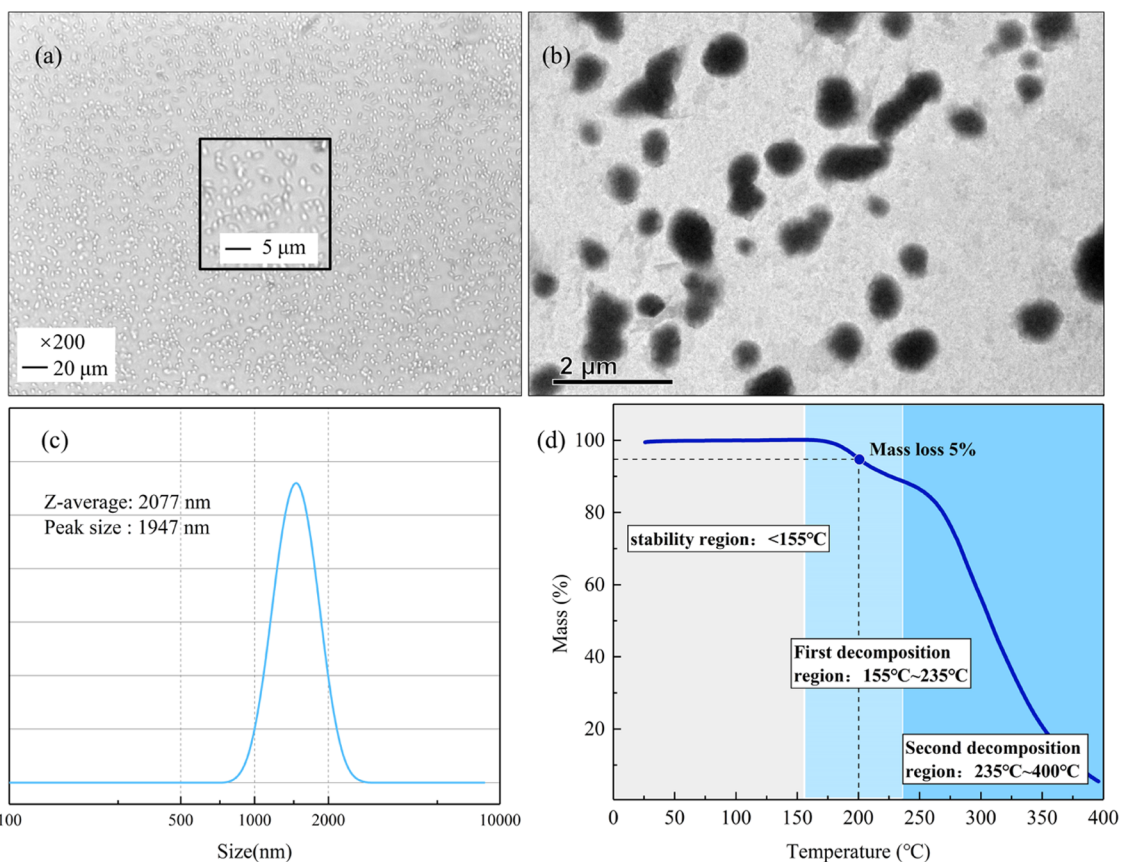
Figure 2. Other characterization results of the product: (a) PM micrograph, (b) TEM micrograph, (c) particle size curve, and (d) TG curve.

was measured by thermogravimetry. In the TG curve (Figure 2d), the product has two-stage thermal decomposition. It is consistent with the thermal decomposition characteristics of the block copolymer. When the mass loss is at 5%, the decomposition temperature is 200 °C. The results show that the product has a strong temperature resistance. It can be used in high-temperature reservoirs.

2.2. Wettability Reversal. Wettability expresses the property for one fluid to adhere to a rock surface in the presence of another immiscible fluid. Therefore, the wettability type controls the distribution of fluids within the rock pore space and framework. Wettability types can be divided into the following: (1) Water-wet: the rock/mineral surface is coated with water, while oil and gas occupy the central position of the largest pores. (2) Oil-wet: the relative positions of oil and water are reversed with respect to the water-wet state; the rock/mineral surface is coated with oil and water is in the center of the largest pores. (3) Intermediate wettability: this term applies to reservoir rocks where there is some tendency for both oil and water to adhere to the pore surface.[28] The wettability of the reservoir can be expressed by the contact angle of the core surface.[29] Figure 3 shows the contact angle and surface free energy of cores treated with different fluoropolymer emulsions. With the increase of concentration, the contact angle between the core and water increased, and the surface free energy of the core decreased. When the fluoropolymer emulsion concentration was 2.0 wt %, the contact angle between the core and water increased from 26° (untreated) to 128°. The surface free energy of the core decreased from 66 to 2.6 mN/m.
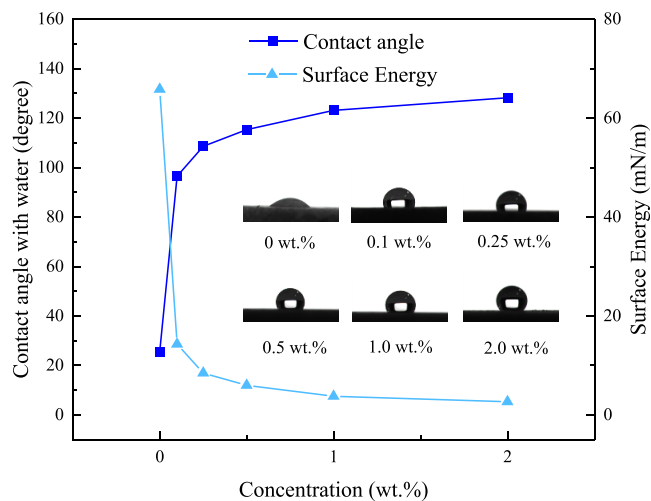
Figure 3. Contact angle and surface free energy of cores treated with different fluoropolymer emulsions.

The contact angle results indicate that the prepared fluoropolymer emulsion has a good wetting reversal ability. After a certain concentration of the emulsion is applied to the reservoir, the wettability of the core can be changed from hydrophilicity to hydrophobicity, and the wettability reversal is achieved.

2.3. Stability. The stability of the emulsion has a qualitative impact on its properties. In general, the emulsion becomes unstable after a certain storage time. The collision of latex particles makes particles bigger and aggregate.[30,31] The stability of the emulsion can be represented by turbidity. The turbidity

of 1 L of water containing 1 mg of SiO_2 is a standard turbidity unit and is denoted 1 NTU. In the experiment, 25 mL of the polymer emulsion was added into a colorimetric tube. Under room temperature and pressure, the turbidity of the emulsion was measured after a period of storage time, and the stability of the emulsion was observed.[32] The turbidity of the emulsion was 557.6 NTU at the initial state. When the emulsion was stored for 30 days, the turbidity of the emulsion increased by less than 2.5% to 571.5 NTU. After the treatment of 2.0 wt % emulsion, the contact angle between the core and water changed from 128 to 123°.

The results show that the fluoropolymer emulsion has good stability and can be stored for a long time at room temperature and pressure. After a certain period of storage, the fluoropolymer emulsion can maintain a good wetting reversal property.

2.4. Imbibition. Generally, the wettability of the reservoir is water wetting. The liquid will be imbibed flowing through the inner channel of the reservoir.[33] The hydrophilic core has a large amount of water imbibition because of the capillary force. Imbibition increases the resistance of liquid migration and reduces the passing capacity of the liquid.[34] Figure 4 shows the
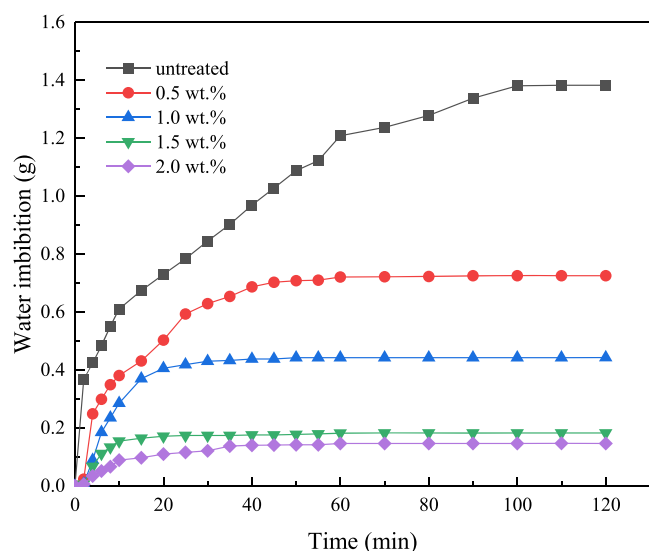
Figure 4. Curves of core imbibition with time.

variation of core imbibition with time. The core imbibition increased with time and finally reached saturation. With the increase of fluoropolymer emulsion concentration, the saturated imbibition capacity of the core decreased. When the concentration was 2.0 wt %, the saturated imbibition of water decreased from 1.38 to 0.15 g, and the saturated imbibition time shortened from 120 to 60 min.

As a result, the fluoropolymer emulsion can reduce the imbibition of the core. The treated core has become hydrophobic. The reversal of reservoir wettability can reduce the dialysis caused by the capillary force so as to reduce the migration resistance of water on the core surface.

2.5. Adsorption. To observe its adsorption, the core surface was tested by scanning electron microscopy (SEM) and energy-dispersive spectroscopy (EDS). In the SEM photograph (Figure 5a,b), it could be clearly observed that the surface of the untreated core was uneven and the exposed rock was angular. After being treated with 2.0 wt % fluoropolymer emulsion, the adsorption layer obviously appeared on the core

surface. The whole surface showed a relatively flat structure, which meant that the roughness of the core surface was improved.

In the EDS results (Figure 5c,d), the element distribution and mass content were obtained. The main elements on the original core were carbon, silicon, oxygen, and common metal elements, which indicated that the core was mainly composed of silica, carbonate, and metal oxides.[35] For comparison, a considerable amount of fluorine element was added into the treated core. As a result, the content of carbon element increased, and the original elements were greatly reduced.

The adsorption of the fluoropolymer leads to the increase of C and F contents. At the same time, it also causes the content change of other elements. What makes the adsorption more intense is the coordination between the ester group in the polymer structure and the metal elements on the core surface. Above all, the fluoropolymer can be effectively adsorbed on the core surface. By changing the chemical structure of the core surface, its roughness is also changed.

2.6. Recovery Rate. The prepared emulsion can change the wettability of the reservoir and reduce the resistance caused by imbibition. The prepared fluoropolymer emulsion can be applied to oil and gas production to enhance the recovery rate.[36] By the visual displacement experiment, the oil recovery rate was tested. In the experiment, we simulated the displacement conditions of the two-stage displacement. First, formation water was injected. Then, after 4 min, hydrogenated polyacrylamide (HPAM) was dissolved in formation water and injected continuously.

In contrast, 2.0 wt % fluoropolymer emulsion replaced the formation water in HPAM displacement. Figure 6 shows the curves of the recovery rate with time. When the fluoropolymer emulsion was 2.0 wt %, the recovery rate increased more than 10% from 58.86 to 69.31%, and the recovery time decreased from 12 to 9 min.

To summarize, the recovery efficiency of crude oil can be effectively improved by adding a certain amount of the fluoropolymer emulsion in the displacement fluid. The application of the fluoropolymer emulsion to oil and gas production can also shorten the recovery time.

3. CONCLUSIONS

In this work, a new type of fluoropolymer microemulsion was prepared by emulsion polymerization. Fluorine-substituted alkyl in the polymer structure can reduce the surface free energy of the interface. The emulsion has micron-size particles and good dispersibility and stability in water, making it play a good role in water-based injection and production fluids. It can also be applied to other fields to change the wettability of the solid−liquid interface.

Fluoropolymers can change the chemical structure of the core surface after adsorption. The wettability of the reservoir changes from hydrophilicity to hydrophobicity. In the application of reservoir reconstruction, the permeability of the reservoir to the water phase is weakened while achieving the inversion of reservoir wettability. Adding a certain concentration of the fluoropolymer microemulsion can enhance the recovery efficiency of crude oil and shorten the recovery time.

There are still some problems to be solved, such as the high cost of fluorine-containing monomers and the limitation of applicable systems. In the later research, the focus of the
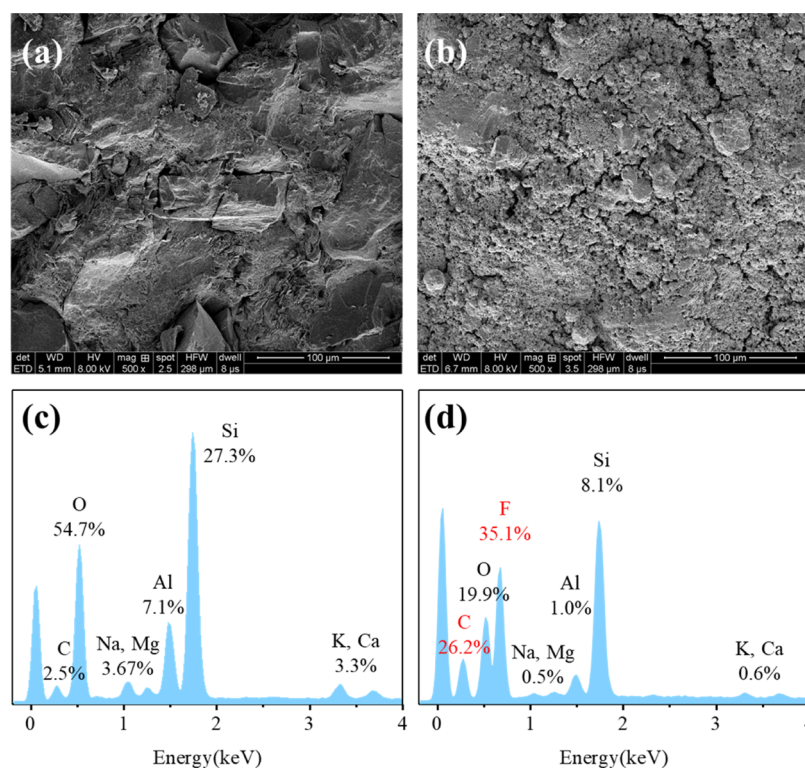
Figure 5. Adsorption and elements of the core surface (SEM: (a) untreated and (b) treated. EDS: (c) untreated and (d) treated).
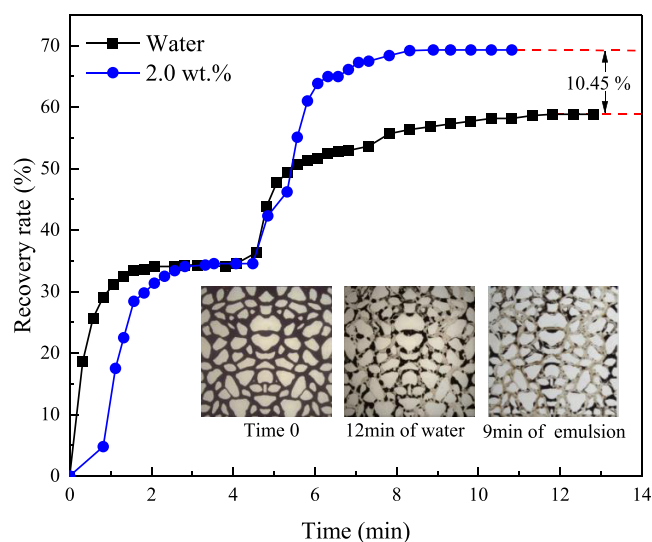
Figure 6. Curves of the recovery rate with time.

research is to reduce the use cost while ensuring the performance of the product.

4. EXPERIMENTAL SECTION

4.1. Materials. The main reagents and samples used in the experiment are listed in Table 1.

4.2. Preparation. Fifty milliliters of distilled water, 1.0 g of MMA (0.01 mol), and 0.1 g of OBS were added into the reaction vessel with condensation and stirring. Then, 4.0 g of F12 (0.01 mol) and 0.1 g of AIBN were dissolved in 50 mL of dimethylformamide (DMF) and added into the reaction vessel and stirred at 60 °C for 15 min under ultrasonic conditions.[37,38] Then, the reaction was heated to 80 °C and continued to react for 6 h. The emulsion with slight white fluorescence was prepared as a fluoropolymer microemulsion (Figure 1b). The reaction scheme is shown in Figure 1a.

4.3. Measurements. *4.3.1. Wettability.* The core was cut into cylinders about 1 cm long and soaked in a certain concentration of the fluoropolymer emulsion. After being soaked at room temperature and pressure for 24 h, the core was taken out and dried at 60 °C for standby.[39] The contact angle between the core surface and water was measured by a

Table 1. Materials in the Experiment

materials	specifications	source contributions
methyl methacrylate (MMA)	AR, 99%	Aladdin Shanghai, China
2,3,4,5,5,5-hexafluoro-2,4-bis(trifluoromethyl)pentyl methacrylate (F12)	AR, 99%	Xeojia Fluorine Silicon Chemical Harbin, China
azodiisobutyronitrile (AIBN)	AR, 99%	Aladdin Shanghai, China
2,5-dimethoxyaniline-4-sulfoanilide (OBS)	AR, 99%	Weng Jiang Reagent Guangdong, China
hydrogenated polyacrylamide (HPAM)	nonionic M_n = 5000 000	Macklin Inc., Shanghai, China
core sample	reservoir shale core K = 10−50 mD	Shengli Oil Field, Sinopec Group Shandong, China
simulated oil	crude oil ρ = 0.8902 g/m³	Shengli Oil Field, Sinopec Group Shandong, China

contact angle instrument (JC2000D, POWEREACH, Shanghai, China).

Based on Young's equation, the surface free energy of the core was calculated by Berthelot's rule[40] according to the following equation

$$\cos \theta = -1 + 2(\gamma_{sv}/\gamma_{lv})^{1/2}$$

In the equation, θ is the contact angle between the core and water (degree), γ_{lv} is the surface free energy of water (72.8 mN/m), and γ_{sv} is the core surface free energy (mN/m).

4.3.2. Imbibition. The core was cut into 5 cm long cylinders and then treated according to the treatment method for measuring the core contact angle. The bottom of the core was in contact with the surface of 50 mL of distilled water vertically, and the top of the core was connected to the analytical balance. The increase of mass with time was recorded as the water imbibition of the core.[41]

4.3.3. Visual Displacement. In the visual displacement experiment, a microglass (5×5 cm^2) was vacuumed and injected with formation water. After the formation water was injected, the simulated oil was injected into the glass model to drive out the formation water. Then, the saturated oil model was obtained (Figure 6, time 0). The oil recovery rate was measured using a visual displacement device (QY-1, HAIAN, Jiangsu, China).

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REFERENCES

(1) Li, Y.; Wang, Y.; Wang, Q.; Liu, Z.; Shi, W.; et al. Achieving the Super Gas-Wetting Alteration by Functionalized Nano-Silica for Improving Fluid Flowing Capacity in Gas Condensate Reservoirs. *ACS Appl. Mater. Interfaces* **2021**, *13*, 10996−11006.

(2) Alvarez, J. O.; Tovar, F. D.; Schechter, D. S. Improving Oil Recovery in the Wolfcamp Reservoir by Soaking/Flowback Production Schedule With Surfactant Additives. *SPE Reservoir Eval. Eng.* **2018**, *21*, 1083−1096.

(3) Lunev, A. S.; Nikitin, A. A.; Shram, V. G.; Afanasov, V. I.; Lysyannikov, A. V.; et al. Influence of Undissolved Gas in Working Fluid on Dynamics of Hydraulic Drive of Equipment for Oil and Gas well Development. *Chem. Pet. Eng.* **2020**, *55*, 963−966.

(4) Aghdam, S. B.; Moslemizadeh, A.; Kowsari, E.; Asghari, N. Synthesis and performance evaluation of a novel polymeric fluid loss controller in water-based drilling fluids: High-temperature and high-salinity conditions. *J. Nat. Gas Sci. Eng.* **2020**, *83*, No. 103576.

(5) Nimana, B.; Canter, C.; Kumar, A. Energy consumption and greenhouse gas emissions in the recovery and extraction of crude bitumen from Canada's oil sands. *Appl. Energy* **2015**, *143*, 189−199.

(6) Hajnos, M.; Calka, A.; Jozefaciuk, G. Wettability of mineral soils. *Geoderma* **2013**, *206*, 63−69.

(7) Yong; Xiao; Shupei; Jianjian; Fang; Hui. Influence of Multiple Factors on the Wettability and Surface Free Energy of Leaf Surface. *Appl. Sci.* **2019**, *9*, 593.

(8) Alnoush, W.; Sayed, A.; Solling, T. I.; Alyafei, N. Impact of Calcite Surface Roughness in Wettability Assessment: Interferometry and Atomic Force Microscopy Analysis. *J. Pet. Sci. Eng.* **2021**, *203*, No. 108679.

(9) Marengo, M.; Antonini, C.; Roisman, I. V.; Tropea, C. Drop collisions with simple and complex surfaces. *Curr. Opin. Colloid Interface Sci.* **2011**, *16*, 292−302.

(10) Wang, Y. C.; Kao, S. H.; Hsieh, H. J. A chemical surface modification of chitosan by glycoconjugates to enhance the cell-biomaterial interaction. *Biomacromolecules* **2003**, *4*, 224−231.

(11) Liu, J.; Yang, T.; Yuan, J.; Chen, X.; Wang, L. Study on Eliminating the Water Blocking Effect in Coal Seams Using Gas-Wetting Reversal Technology. *ACS Omega* **2020**, *5*, 30687−30695.

(12) Li, Y.; Wang, Y.; Qian, W.; Tang, L.; Lin, Y.; Wang, G.; Ren, Z. Optimization the synthesis parameters of gas-wetting alteration agent and evaluation its performance. *Colloids Surf., A* **2018**, *558*, 438−445.

(13) Siegbahn, P.; Pettersson, L.; Wahlgren, U. A theoretical study of atomic fluorine chemisorption on the Ni(100) surface. *J. Chem. Phys.* **1991**, *94*, 4024−4030.

(14) Vidal, R.; Ferron, J.; Meyer, C. I.; Riascos, V. Q.; Bonetto, F. The influence of surface defects on the low energy scattering of Ar ions from a Cu(111) surface. *Surf. Sci.* **2019**, *690*, No. 121482.

(15) Churchley, D.; Rees, G. D.; Barbu, E.; Nevell, T. G.; Tsibouklis, J. Fluoropolymers as low-surface-energy tooth coatings for oral care. *Int. J. Pharm.* **2008**, *352*, 44−49.

(16) Kichigina, G. A.; Bol'Shakov, A. I.; Kushch, P. P.; et al. Radiation-initiated synthesis of tetrafluoroethylene telomers in 1,2-dibromotetrafluoroethane at a constant pressure of the monomer in the reactor. *Russ. J. Appl. Chem.* **2014**, *87*, 518−524.

(17) Son, T. Y.; Ae, J. M.; Yong, N. S. Amorphous Fluoropolymer Membrane for Gas Separation Applications. *J. Nanosci. Nanotechnol.* **2018**, *18*, 6206−6212.

(18) Mikhaylov, V. G.; Ponomarev, A. I.; Topolnikov, A. S. Prediction of gas factor taking into account gas dissolved in the water at late stages development of oil fields. *SOCAR Proc.* **2017**, *3*, 41−48.

(19) Zhu, J.; Liu, S.; Zhu, N.; Ye, D. *The Novel Energy Policy Evaluation Method and Its Application in Oil and Gas Fields in China*; Springer: Berlin Heidelberg, 2010.

(20) Khapli, S.; Jagannathan, R. Supercritical CO_2 based processing of amorphous fluoropolymer Teflon-AF: Surfactant-free dispersions and superhydrophobic films. *J. Supercrit. Fluids* **2014**, *85*, 49−56.

(21) Koroleva, M. Y.; Yurtov, E. V. Water Transport by Nanodispersion Droplets in a Water-in-Oil Emulsion. *Colloid J.* **2003**, *65*, 35−39.

(22) Wang, Y.; Liang, L.; Li, Y.; Liu, B.; Tang, L. Preparation and application of a fluoropolymer emulsion as novel wettability reversal agent. *Colloids Surf., A* **2020**, *612*, No. 125985.

(23) Jin, J.; Wang, H.; Jing, Y.; Liu, M.; Wang, D.; Li, Y.; Bao, M. An efficient and environmental-friendly dispersant based on the synergy of amphiphilic surfactants for oil spill remediation. *Chemosphere* **2019**, *215*, 241−247.

(24) Jhnigen, S.; Sebastiani, D. Carbon Atoms Speaking Out: How the Geometric Sensitivity of 13C Chemical Shifts Leads to Understanding the Colour Tuning of Phycocyanobilin in Cph1 and AnPixJ. *Molecules* **2020**, *25*, No. 5505.

(25) Dong, H.; Lee, S. Y.; Yi, G. R. Controlling the size and surface morphology of carboxylated polystyrene latex particles by ammonium hydroxide in emulsifier-free polymerization. *Macromol. Res.* **2009**, *17*, 397−402.

(26) Huppertz, H.; Kotzyba, G.; Hoffmann, R. D.; Pöttgen, R. Decomposition of EuPdIn and EuPtIn at high temperature and high pressure-formation of the hexagonal Laves phases EuPd0.72In1.28 and EuPt0.56In1.44. *J. Solid State Chem.* **2002**, *169*, 155−159.

(27) Xiong, G.; Zhu, W. Coupling effects of high temperature and pressure on the decomposition mechanisms of 1,1-diamino-2,2-dinitroehethe crystal: Ab initio molecular dynamics simulations. *J. Chin. Chem. Soc.* **2020**, *67*, 1571−1578.

(28) Jürgen, H. S. Pore Space Properties. *Dev. Pet. Sci.* **2015**, *65*, 21−84.

(29) Stammitti-Scarpone, A.; Acosta, E. J. Solid-Liquid-Liquid Wettability of Surfactant-Oil-Water Systems and Its Prediction around the Phase Inversion Point. *Langmuir* **2019**, *35*, 4305−4318.

(30) Urbina-Villalba, G.; Lozsán, A.; Rahn, K.; Romero-Cano, M. S. Calculation of the stability ratio of suspensions using Emulsion Stability Simulations. *Comput. Phys. Commun.* **2009**, *180*, 2129−2139.

(31) Mukhtar, N. A. M.; Aziz, A.; Hagos, F. Y.; Noor, M. M.; Awalludin, M. The Influence of Formulation Ratio and Emulsifying Settings on Tri-Fuel (Diesel-Ethanol-Biodiesel) Emulsion Properties. *Energies* **2019**, *12*, 1708.

(32) Bouchaou, L.; Mangin, A.; Chauve, P. Turbidity mechanism of water from a karstic spring: example of the Ain Asserdoune spring (Beni Mellal Atlas, Morocco). *J. Hydrol.* **2002**, *265*, 34−42.

(33) Gahrooei, H. R. E.; Ghazanfari, M. H. Application of a water based nanofluid for wettability alteration of sandstone reservoir rocks to preferentially gas wetting condition. *J. Mol. Liq.* **2017**, *232*, 351−360.

(34) Fang, C.; Qiao, R. Surface hydration drives rapid water imbibition into strongly hydrophilic nanopores. *Phys. Chem. Chem. Phys.* **2017**, *19*, 20506−20512.

(35) Jja, B.; Jsab, C.; Kla, B.; Xha, B.; Jwa, B.; Jla, B.; Yba, B.; Xuan, G. A.; Jz, A.; Jia, L. A. Magnetic-responsive CNT/chitosan composite as stabilizer and adsorbent for organic contaminants and heavy metal removal. *J. Mol. Liq.* **2021**, *334*, No. 116087.

(36) Jin, J.; Sun, J.; Rong, K.; Lv, K.; Wang, J.; et al. Gas-Wetting Alteration by Fluorochemicals and Its Application for Enhancing Gas Recovery in Gas-Condensate Reservoirs: A Review. *Energies* **2020**, *13*, No. 4591.

(37) Li, Y.; Wang, Y.; Jin, J.; Wang, K.; Tang, L.; Zhang, Y.; Wang, G.; Dai, C. Synthesis and Evaluation of Two Gas-Wetting Alteration Agents for a Shale Reservoir. *Energy Fuel* **2018**, *32*, 1515−1524.

(38) Yw, A.; Yl, A.; Qian, W. B.; Lt, A.; Lei, L. A.; Yz, C.; Jl, A.; Ning, X. A. The influence of fluorochemical-modified graphene oxide on the gas-wetting alteration of reservoir cores. *Colloids Surf., A* **2021**, *620*, No. 126565.

(39) Wang, Y.; Li, Y.; Qian, W.; Lei, L. A.; Zhang, C.; Meng, L.; Tang, L.; Lan, J.; Jiang, B. Super gas-wetting alteration of reservoir cores using fluorine-modified nanocrystalline cellulose. *J. Mol. Liq.* **2021**, *333*, No. 115933.

(40) Girifalco, L. A.; Good, R. J. A theory for the estimation of surface and interfacial energies. I. Derivation and application to interfacial tension. *J. Phys. Chem. A* **1957**, *61*, 904−909.

(41) Sun, J.; Chang, X.; Zhang, F.; Bai, Y.; Lv, K.; Wang, J.; Zhou, X.; Wang, B. Salt-Responsive Zwitterionic Polymer Brush Based on Modified Silica Nanoparticles as a Fluid-Loss Additive in Water-Based Drilling Fluids. *Energy Fuel* **2020**, *34*, 1669−1679.