



The Effect of Formation Water Salinity on the Minimum Miscibility Pressure of CO₂-Crude Oil for Y Oilfield

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CO₂ miscible flooding is an important technology for enhancing oil recovery and greenhouse gas storage in the world. As a tertiary recovery technology, it is usually applied after water flooding. Therefore, the actual reservoirs usually contain a lot of injected water in addition to connate water. The salinity of these formation waters varies from place to place. CO₂ is an acid gas. After it is injected into the reservoir, it easily reacts with formation water and rock and affects the physical properties of the reservoir. However, no research results have been reported whether this reaction affects the minimum miscibility pressure (MMP) of CO₂-crude oil, a key parameter determining miscible flooding in formation water. Based on CO₂-formation water-rock interaction experiments, this paper uses the core flooding method to measure the CO₂-crude oil MMP under different salinity in formation water. Results show that CO₂ causes a formation water pH decrease from 7.4 to 6.5 due to its dissolution in formation water. At the same time, CO₂ reacts with formation water, albite, potassium feldspar, and carbonate minerals in the cores to generate silicate and carbonate precipitates, which could migrate to the pore throat together with the released clay particles. Overall, CO₂ increased core porosity by 5.63% and reduced core permeability by 7.43%. In addition, when the salinity of formation water in cores was 0, 4,767, and 6,778 mg/L, the MMP of CO₂-crude oil was 20.58, 19.85, and 19.32 MPa, respectively. In other words, the MMP of CO₂-crude oil decreased with the increase of salinity of formation water.

Keywords: CO₂ miscible flooding, minimum miscibility pressure, formation water salinity, core displacement experiment, CO₂-formation water-rock interaction experiment

INTRODUCTION

CO₂ flooding is an effective method to improve oil recovery. There are two displacement modes: CO₂ miscible flooding and CO₂ immiscible flooding, depending on whether the reservoir pressure can reach the minimum miscible pressure (MMP) of CO₂-crude oil (Zhang et al., 2019; Syed et al., 2020; Han et al., 2018; Jishun et al., 2015). CO₂ miscible flooding can result in much higher oil recovery than CO₂ immiscible flooding because the miscible condition can reduce capillary pressure to zero, and residual oil can be significantly reduced in flooded areas/zones. Therefore, MMP, which determines whether the reservoir oil and CO₂ are miscible or not, is particularly important.

At present, studies on the influencing factors of MMP mainly focus on CO₂ purity (Cumicheo et al., 2014; Zhao et al., 2020; Cumicheo et al., 2014), oil composition (Dong et al., 2001; Hemmati-Sarapardeh et al., 2014; Yalcin and Faruk, 2020), temperature, and pressure (Zolghadr et al., 2013;

TABLE 1 | Compositional results of the oil sample.

Carbon no.	mol%	wt%	Carbon no.	mol%	wt%
C ₁	46.35	6.16	C ₁₆	1.74	2.46
C ₂	5.42	1.56	C ₁₇	0.82	1.73
C ₃	3.35	1.32	C ₁₈	0.67	2.41
C ₄	1.88	0.92	C ₁₉	0.78	2.02
C ₅	1.26	0.74	C ₂₀	0.83	2.05
C ₆	2.85	1.32	C ₂₁	1.36	4.13
C ₇	3.42	1.42	C ₂₂	0.31	3.86
C ₈	3.96	3.11	C ₂₃	0.36	4.02
C ₉	3.31	2.76	C ₂₄	0.24	2.83
C ₁₀	1.95	2.31	C ₂₅	0.41	1.85
C ₁₁	2.36	1.46	C ₂₆	0.3	1.49
C ₁₂	2.86	2.68	C ₂₇	0.23	0.89
C ₁₃	2.05	2.97	C ₂₈	0.15	0.74
C ₁₄	0.71	1.42	C ₂₉	0.34	1.66
C ₁₅	2.35	2.55	C ₃₀₊	7.38	35.16

Jamiu, 2013; Jia et al., 2019; Moeini et al., 2014). In fact, the reservoir is usually flooded by water before CO₂ injection, and inevitably, there is connate and injected water with different salinity in formation. As a highly active gas, CO₂ easily reacts with formation water after being injected into a reservoir (Ampomah et al., 2017; Ren et al., 2019). It can impact the physico-chemical balance between reservoir rock and formation water and result in a change in the reservoir rock's physical properties. However, no research has been reported about whether and how the interactions affect the MMP for the formations with different water salinity.

Both theoretical calculation and laboratory experimental methods have been used to obtain the MMP of CO₂-crude oil (Zendehboudi et al., 2013; Ahmadi et al., 2015; Ahmadi et al., 2017; Ahmadi et al., 2018). Theoretical calculation methods mainly include empirical formulae (Yellig and Metcalfe, 1980), multiple contacts (Adekunle and Hoffman, 2016), numerical simulations (Ekechukwu et al., 2020), and equation of states (Karkevandi-Talkhoonchek et al., 2017), etc., because the application range of different calculation methods is different, and the results are greatly affected by parameters. The experimental measurement methods mainly include the slim tube test (STT) (Flock and Nouar, 1984; Mogensen, 2016), rising bubble apparatus (RBA) (Christiansen and Haines, 1987; Novosad et al., 1990; Czarnota et al., 2017), vanishing interfacial tension technique (VIT) (Rao, 1997; Saeedi Dehaghani and Soleimani, 2020), X-ray computerized scanner (Liu et al., 2015; Ahmad et al., 2016), magnetic resonance imaging technique (MRI) (Liu et al., 2016; Karkevandi-Talkhoonchek et al., 2018), core displacement experiment (Zhang and Gu, 2015; Ennin and Grigg, 2016), and so on. Among these methods, STT is the most common method that has been widely used because it usually has a long tube packed with sand or glass beads and can effectively simulate the fluid flow in porous media. However, core tubing cannot represent the mineral composition, porosity, permeability, and connate water saturation of real reservoir rocks. Therefore, it cannot be used for our study because our goal is to investigate formation the water salinity effect on the MMP of CO₂-crude oil in reservoirs. In this paper, we use CO₂-

formation water-rock interaction experiments to reveal their interaction mechanisms using reservoir rock samples. On the basis of these experiments, MMP is measured using core displacement experiments in the formation water with different salinity. The results are of great significance to the optimization design of CO₂ miscible flooding.

EXPERIMENTS

Materials

The oil samples were taken from Y oilfield. Their components and related properties are shown in **Tables 1, 2**, respectively, at reservoir pressure (22.05 MPa) and temperature (98.5°C).

Distilled water and synthetic formation water from different areas of Y oilfield were used for the experiments. The composition and salinity of the formation waters are shown in **Table 3**. The pH values of the two formation waters were 7, 7.4, and 7.8, respectively. CO₂ with purity of 99.99% was provided by United Gas Ltd. for CO₂-water-rock interaction and core displacement tests.

The wettability of four natural cores from Y oilfield is water wet. The length, diameter, porosity, and permeability of natural cores were 30 cm, 2.5 cm, 23%, and $50 \times 10^{-3} \mu\text{m}^2$, respectively. The core minerals are mainly composed of 53% quartz, 32% feldspar (plagioclase, potassium feldspar), and 15% lithic (clay minerals, a small amount of carbonate minerals). The carbonate minerals are mainly calcite and ankerite.

Experimental Apparatus

The core flooding apparatus shown in **Figure 1** was used to investigate the CO₂-water-rock interaction and obtain MMP. The apparatus is composed of an ISCO pump (260D, Teledyne ISCO Inc., United States), core holder (40 MPa), backpressure regulator, pressure gauges (OMEGA Engineering INC., Canada) with an accuracy of ± 0.01 MPa, accumulators (40 MPa), and an oven. Both the accumulator and core holder were put in the oven to simulate reservoir temperatures. The changes in the porous throat before and after the CO₂-water-rock reaction were observed by Nano Voxel-4000 CT scanner (Sanying Precision Instrument Inc., China). The PH value of the produced water was tested by a PHB-4 pH detector (Sinameasure AOTumation Technology Inc., China).

Experimental Procedures

CO₂-formation Water-Rock Interaction Experiment

Through the CO₂-formation water-rock interaction experiment, the reaction mechanism of gas, water, and rock is revealed. The procedures are listed as follows in detail:

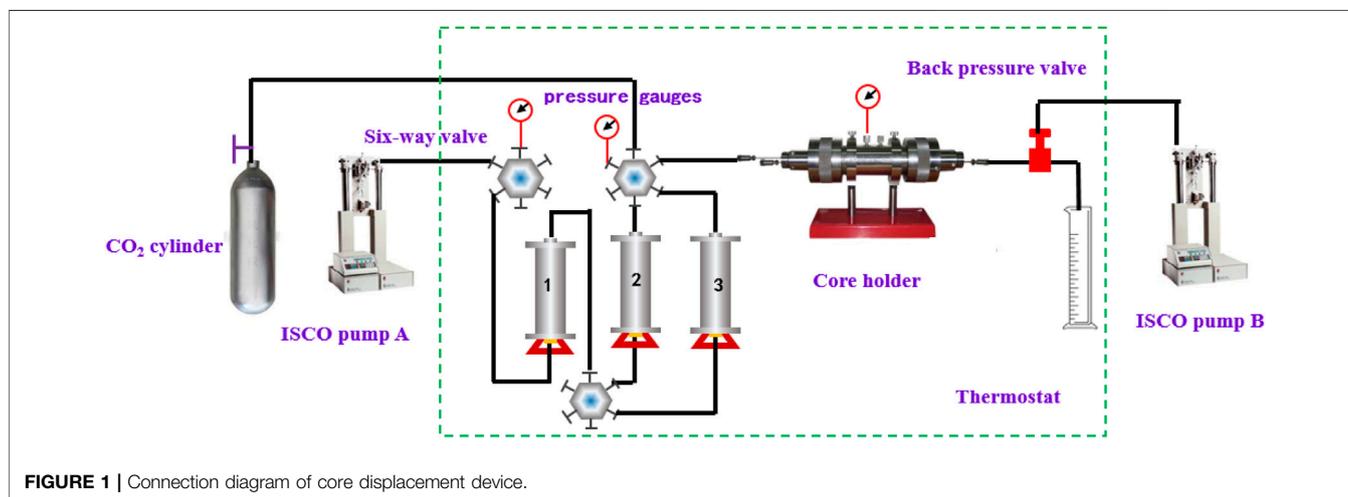
- 1) To compare the changes of core physical properties before and after displacement, the end face of the core is cut. The remaining core is put into the core holder or vacuum and then saturated with formation water.
- 2) CO₂ and formation water are injected into the container through the ISCO pump A, and the formation water is in the supersaturated state during the whole experiment.

TABLE 2 | Physical properties of the oil sample.

Parameter	Gas oil ratio (m ³ /m ³)	Volume ratio of formation oil (m ³ /m ³)	Saturation pressure (MPa)	Density of formation oil (g/cm ³)	Formation oil volume factor (mPa·s)
Value	22.8	1.106	4.95	0.9377	3.62

TABLE 3 | Compositions of formation water.

Compositions	NaHCO ₃	NaCl	KCl	MgSO ₄	Na ₂ SO ₄	CaCl ₂	Total
Formation Water 1	2,718	1789	20	62	114	64	4,767
Formation Water 2	3,866	2,545	28	88	161	90	6,778

**FIGURE 1** | Connection diagram of core displacement device.

- 3) The formation water is injected into the core at a flow rate of 0.3 ml/min. Effluent samples are taken at the outlet every 10 min. The pH value and ion concentration of these samples are measured by the PHB-4 pH detector and DiONEX 500 chromatograph.
- 4) After the core flooding test, the core is put into the oven for drying. When the mass of the core is no longer changed, the physical parameters of the core are measured, and the core and the original undergo CT scan.

CO₂ Miscible Flooding Experiment

The CO₂ miscible flooding experiment is used to measure the CO₂-crude oil MMP in the formation water with different salinity. The detailed procedures are as follows:

- 1) The core is placed into the core holder and vacuumed; then it is saturated with distilled water by manual pump.
- 2) The core is placed into the core holder and vacuumed; then it was saturated with distilled water by manual pump.
- 3) The pressure of the backpressure valve is set by ISCO pump B, and its value is below the injection-end pressure of 0.8–1 MPa. A constant pressure flow of 5 MPa is set by ISCO pump A.

Then, the displacement experiment is carried out. When the volume of cumulative injection is greater than 1.5 PV, the displacement is stopped, and the oil recovery at this pressure is calculated.

- 4) The displacement experiment at the next pressure is carried out. CO₂ pressures were 5, 10, 15, 20, 25, 27, and 30 MPa. The pressure in the CO₂ cylinder is 5 MPa. Displacement pressure is adjusted by two containers.
- 5) After completion of the experiments at each pressure, the core and formation water were replaced to carry out the next experiment.

RESULTS AND DISCUSSION

Changes of pH and Ion Concentration of Produced Liquid

CO₂ is a kind of acidic gas, which easily reacts with formation water after being injected into the reservoir, breaking the physical-chemical balance between rock and formation water and then affecting the core physical parameters. To reveal the law of CO₂-water-rock interaction, the PH value of the produced liquid and the concentration of related ions were measured.

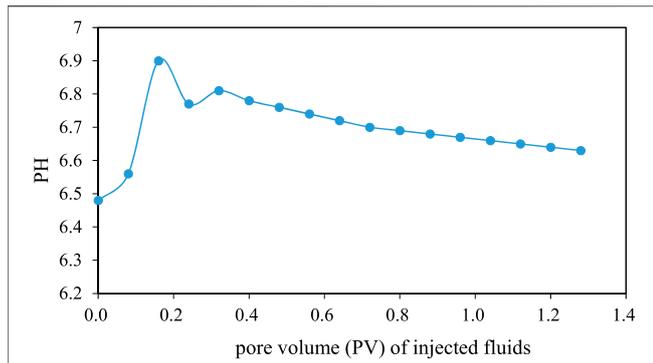


FIGURE 2 | Curve of pH values of produced water over the PV.

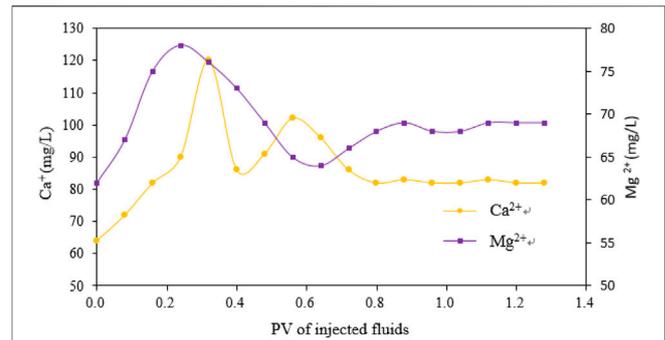


FIGURE 4 | Curve of Ca²⁺ and Mg²⁺ concentration of produced water over the PV.

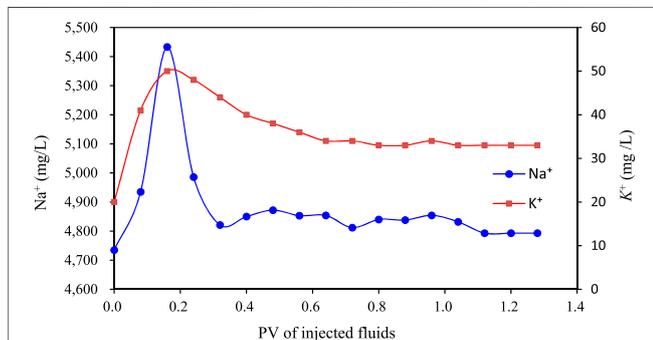


FIGURE 3 | Curve of Na⁺ and K⁺ concentration of produced water over the PV.

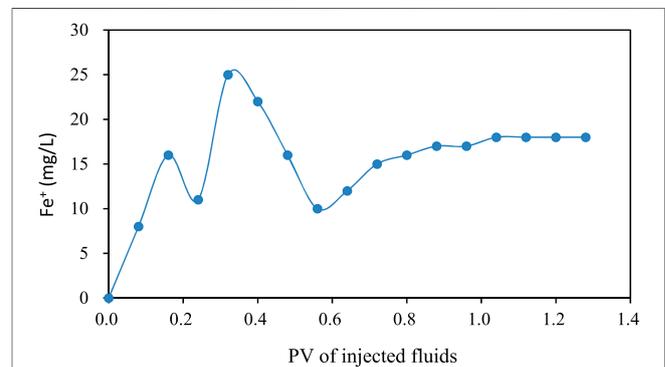


FIGURE 5 | Curve of Fe²⁺ concentration of produced water over the PV.

The test results of PH values of the produced liquid are shown in **Figure 2**. When the formation water is saturated with CO₂ in the container, the formation water is weakly acidic, and the pH value decreases from 7.4 to 6.48. After the formation water was injected into the core, the pH value of the solution increased significantly at the beginning of the experiment, and then decreased to a stable level with the increase of the injected PV. The analysis is as follows: The main reason for the formation water changing from alkaline to acidic is that CO₂ acidifies the water and forms free H₂CO₃. The reaction equation is as follows:



The variation curve of pH values with injected PV suggests that the reactions occurred between core, water, and CO₂, which significantly reduced the content of H⁺ in the above equation, and then the pH value of the solution slowly decreased to a stable level as the reaction reached equilibrium.

Figure 3 shows the changed Na⁺ and K⁺ concentrations of produced water with the injected PV. The mass concentrations of Na⁺ and K⁺ increased first and then decreased to a stable level with the increase of injected PV. The chemical reactions between CO₂, core, and water depend on the mineral compositions of the core. The known mineral

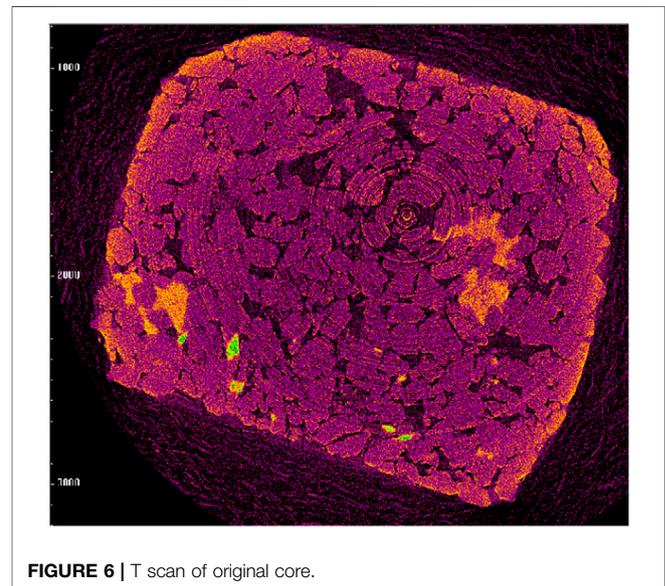


FIGURE 6 | T scan of original core.

compositions of the cores are mainly quartz, plagioclase, potassium feldspar, clay minerals, and carbonate minerals (ankerite and calcite). The change of mass concentrations of

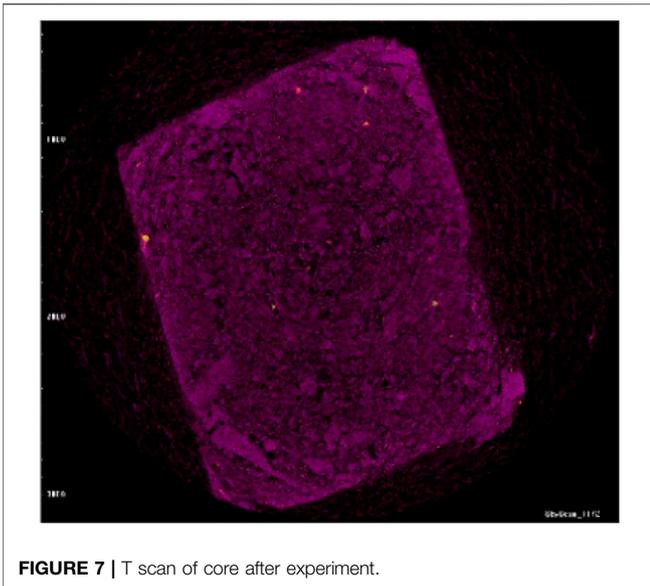
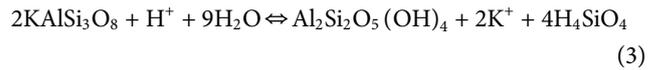
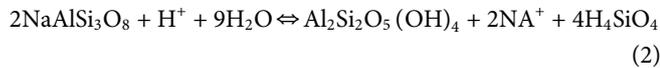


FIGURE 7 | T scan of core after experiment.

Na⁺ and K⁺ in produced effluent samples indicates that albite (NaAlSi₃O₈) and potassium feldspar (KAlSi₃O₈) in the core reacted with acidic formation water. The reaction equation is as follows:



These reactions generated silicate minerals and released Na⁺ and K⁺, leading to a significant increase in their mass concentration at the beginning of the experiment. With the progress of the experiment, albite (NaAlSi₃O₈) and potassium feldspar (KAlSi₃O₈) reached solve-precipitation equilibrium, and the ion concentration decreased to a stable level.

Figures 4, 5 show the changes of Mg²⁺, Ca²⁺, and Fe²⁺ concentrations. The concentrations of Mg²⁺, Ca²⁺, and Fe²⁺ increased first and then fluctuated to a stable level with the increase of injected PV. The change of the mass concentration of Mg²⁺, Ca²⁺, and Fe²⁺ in the reaction solution was mainly related to the dissolution and precipitation of calcite and ankerite. At the beginning of the experiment, the concentrations of Ca²⁺, Fe²⁺, and Mg²⁺ increased, which indicated that carbonate minerals (calcite and ankerite) were corroded by the CO₂ acid solution. The reaction consumes H⁺ in the solution, which leads to the increase of pH value in the solution. The calcite dissolution equation of the solution is shown in Eq. 4, and the ankerite dissolution equation is shown in Eq. 5.

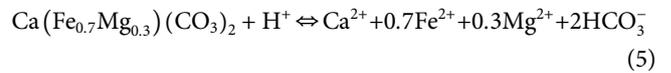


TABLE 4 | Changes of core porosity and permeability before and after the experiment.

Porosity(%)		Rate of change in porosity(%)	Permeability(x10 ⁻³ μm ²)		Rate of change in permeability(%)
Before the experiment	After the experiment		Before the experiment	After the experiment	
21.3	22.5	5.63	20.2	18.7	-7.43

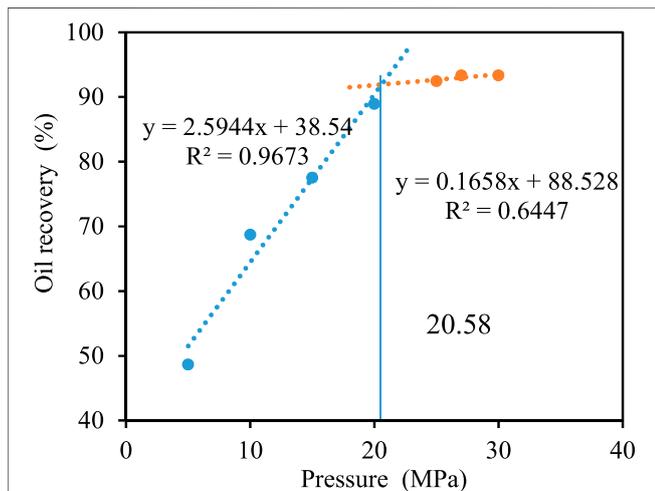


FIGURE 8 | Oil recovery under different displacement.

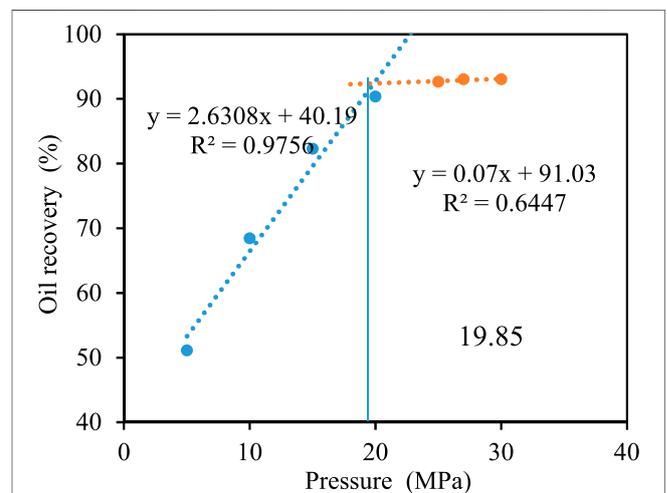
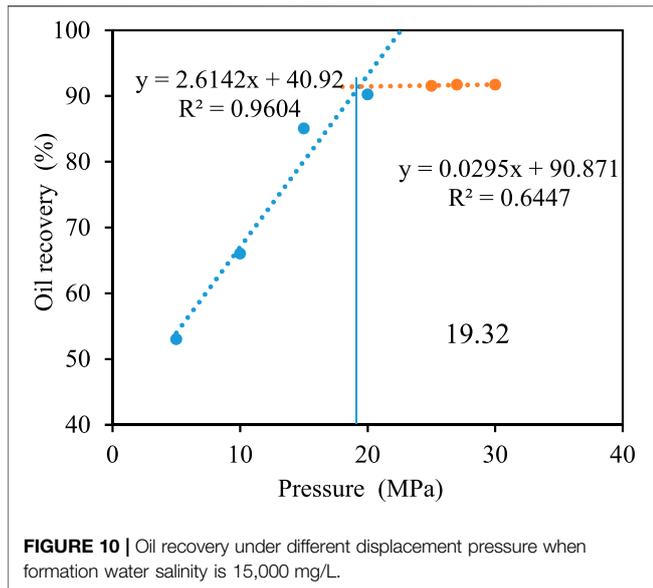
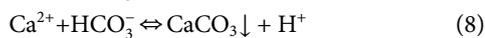
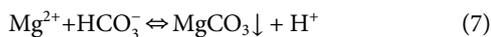
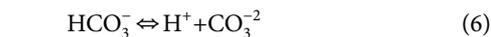


FIGURE 9 | Oil recovery under different displacement. Pressure when formation water salinity is 0 mg/L pressure when formation water salinity is 6778 mg/L.



At the same time, with the progress of the experiment, the above reaction promoted the continuous ionization of H₂CO₃ and lowered the PH value of the solution. Meanwhile, some cations in the solution, such as Mg²⁺ and Ca²⁺, react with bicarbonate to form insoluble carbonate. This reaction released a large amount of H⁺, which reduced the concentration of metal cations in the solution. The reaction formulas are shown in Eq. 1, Eqs 6–8:



Changes of Core Physical Properties

Figures 6, 7 show the CT scanning results of natural cores before and after the experiment. The pore volume of the core increases obviously after displacement. This is consistent with the core

porosity measurement results after the experiment (Table 4). However, the core permeability decreases obviously after displacement. The relevant analysis is as follows: The formation water filled with saturated Ca²⁺ dissolves the core. At the same time, it generates corresponding precipitates (Al₂Si₂O₅(OH)₄, H₄SiO₄), and a lot of Mg²⁺ and Ca²⁺. With the increase of the concentration of these divalent ions, they react with the bicarbonate in the formation water to generate MgCO₃ and CaCO₃ precipitates. The dissolution of these new mineral phases with carbonate cements releases clay particles that were transported by fluid to the pore throats, where the pores were blocked and the permeability of the core was reduced.

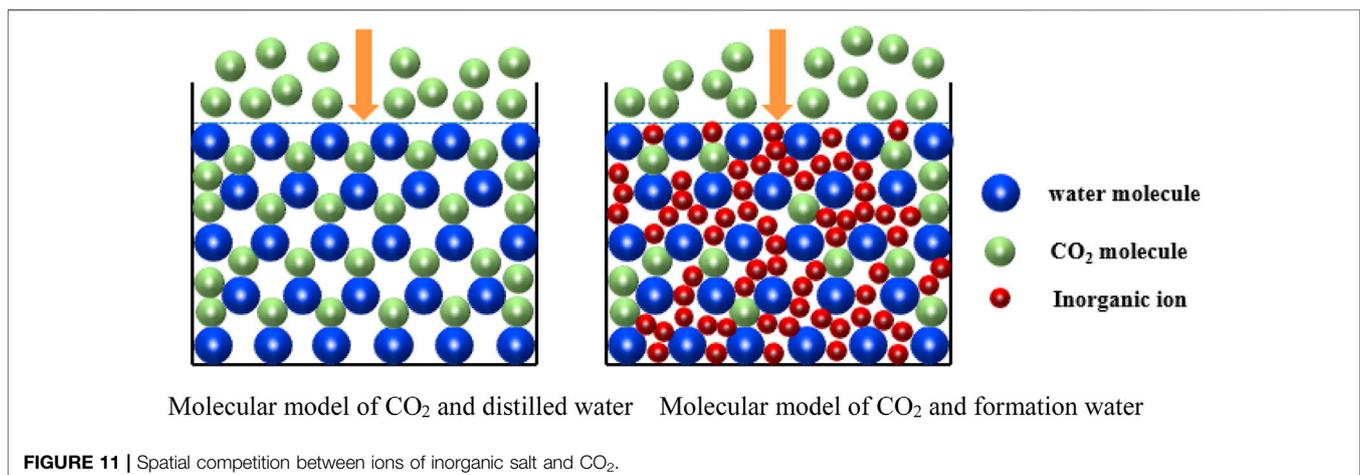
CO₂-Crude Oil MMP Under Formation Water of Different Salinity

To reveal the influence of formation water salinity on the MMP of CO₂ and crude oil, the core displacement method is used to measure the MMP under different formation water. It can be seen from Figures 8–10 that the MMP is 20.80, 19.85, and 19.21. The MMP of CO₂-crude oil decreases. The analysis is as follows:

There is a spatial competition effect in the dissolution of CO₂ in formation water. Compared with CO₂, the ionic radius of inorganic salt is smaller, and it more easily fills the gap in water molecules (Figure 11). This reduces the dissolution space for CO₂, resulting in less CO₂ soluble in water and more CO₂ soluble in crude oil in the core saturated with high salinity, resulting in more miscibility. Therefore, the higher the salinity of the formation water, the smaller the MMP between CO₂ and crude oil.

When the pressure is greater than 7.38 MPa, CO₂ reaches supercritical state. Under the action of its extraction and pore-throat capillary force, NaCl in the formation water is pumped to the mineral surface. The precipitated NaCl crystals with mineral precipitation of CO₂-water-rock interaction are carried by CO₂ for blocking the pore throat.

This increases the pore volume and reduces the permeability of the core. The change of the physical properties of the core increase the gas breakout time and



make the compositional exchange between CO₂ and crude oil more intense in the smaller pore space. Thus, the MMP between CO₂ and crude oil is lower.

Molecular model of CO₂ and distilled water Molecular model of CO₂ and formation water.

CONCLUSION

Based on CO₂-formation water-rock interaction and core displacement method experiments, the following conclusions are drawn in this paper:

1)CO₂ can react with formation water to decrease the formation water pH from 7.4 to 6.5.

2) CO₂ can react with minerals in the core to form insoluble silicate and carbonate precipitates. These new minerals migrate to the pore throat together with the released clay particles, resulting in the increase of the core porosity by 5.63% and the decrease of the core permeability by 7.43%.

3)Formation water salinity influences MMP. MMP of CO₂-crude oil decreased with the increase of formation water salinity in the cores.

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DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

YP conceived and designed the analysis JL wrote the paper and performed the analysis CL collected the data ZL collected the data.

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Conflict of Interest: XG was employed by the China United Coalbed Methane Co., Ltd. CL was employed by CNOOC Safety Technology Service Co., Ltd. Shanghai Branch.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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