Microscopic Oil Displacement Mechanism of CO₂ in Low-Permeability Heterogeneous Glutenite Reservoirs in the Junggar Basin

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ABSTRACT: Carbon dioxide (CO₂) flooding is a promising method for developing low-permeability heterogeneous glutenite reservoirs (LPHGRs) featured with low natural energy. Herein, the focus of this work was to study the microscopic oil displacement mechanism of CO₂ in LPHGRs. First, the micropore structure and mineral composition of LPHGRs were analyzed, and the effect of CO₂ on low-permeability reservoirs was then studied. Also, the mechanism of CO₂ displacement in low-permeability reservoirs on a pore scale was analyzed using nuclear magnetic resonance technology in different dimensions. The experimental results showed that the mineral composition of the rock mainly included quartz, feldspar, and clay minerals. The core pores were poorly developed and highly heterogeneous. The clay and other mineral particles produced by the dissolution reaction of rocks and minerals migrated and deposited with formation fluid to damage the reservoir. On the contrary, it also improved the pore structure, increased pore space, and increased reservoir permeability. The miscible interaction between CO₂ and crude oil formed a stable displacement front, which enabled crude oil to be well displaced from the macropores and medium pores. However, this interaction generated negligible effects on the small pores. The experimental results provided important indicators for CO₂ development in LPHGRs.

1. INTRODUCTION

Low-permeability reservoirs account for a large proportion of the world’s reservoir types, which are rich and widely distributed, mainly in Canada, the USA, Mexico, and China. Similarly, this kind of reservoirs account for a great percentage in China, with proven reserves of 14.1 billion tons, accounting for 49.2% of the total proven reserves. The remaining oil of low-permeability reservoirs in China is 43.1 billion tons, accounting for roughly 60% of the total remaining oil.

The natural energy of glutenite reservoirs is low. During the initial stage of reservoir development, crude oil recovery mainly depends on the natural energy of the reservoir itself. The natural energy of conglomerate mainly includes elastic energy and dissolved gas energy. Generally, the recovery rate depending on natural energy is about 10%. Due to low natural energy, the pressure of the reservoir decreases significantly during development. Reservoir energy needs to be replenished to restore formation pressure.

For conventional reservoirs, water flooding was often used, but for low-permeability reservoirs, water flooding would face many unfavorable factors, such as high injection pressure, the compatibility between water and formation rocks, and so forth. Therefore, gas flooding had been applied in low-permeability reservoirs, in which CO₂ had become a key strategy for the development of these reservoirs. The main way it enhanced oil recovery was by dissolving in the oil phase to expand the volume and lower the viscosity of oil.

CO₂ as a greenhouse gas was a product of industrial production and human activities, which produced a serious impact on the global climate. Geological storage of CO₂ was widely regarded as a good and important method to reduce carbon emissions. Simultaneously, the storage of CO₂ could be used for reservoir development. As reported, the mechanisms of CO₂ development in low-permeability reservoirs could be summarized as expanding crude oil, reducing crude oil viscosity, dissolving carbonate cement in oil layer, dissolved gas flooding, improving the oil–water flow ratio, reducing interfacial tension, forming miscible fluid, and so forth.

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For low-permeability reservoirs, many peers had also conducted some studies regarding the effect of CO₂ on enhancing oil recovery. Xiao et al.²⁰ claimed that compared to water flooding, CO₂ injection could improve oil displacement efficiency in micropores significantly. Feng et al.²¹ evaluated the miscibility of crude oil and CO₂ in low-permeability reservoirs. The results indicated that miscibility could significantly increase production of low-permeability reservoirs. Kun et al.²² found that CO₂ injection after water flooding development also effectively enhances crude oil recovery from low-permeability reservoirs. Nan et al.²³ concluded that compared with nitrogen (N₂) and hydrocarbon gas, CO₂ owned lower miscibility pressure, delayed gas breakthrough, and better development effects. All these studies proved that CO₂ played a very important role in the development of low-permeability reservoirs.

This paper studied the microscopic oil displacement mechanism of CO₂ in the low-permeability heterogeneous glutenite reservoirs (LPHGRs) in the Junggar Basin, China. First, the interaction between crude oil and CO₂ in the low-permeability reservoir was studied. Then, the micropore structure and mineral composition of the low-permeability reservoir was analyzed by scanning electron microscopy (SEM) and X-ray diffraction, and the effect of CO₂ on the low-permeability reservoir was also studied. Finally, the mechanism of CO₂ displacement on a pore scale was analyzed using nuclear magnetic resonance (NMR) technology in different dimensions. The research results had an in-depth understanding for the development of CO₂ injection used to exploit LPHGRs.

2. MATERIAL AND METHODS

2.1. Material. 2.1.1. Crude Oil. The crude oil sample used in the experiment was taken from the Junggar Basin of Xinjiang, China. 7890B GC (Agilent Technologies Inc., USA) was used to analyze the carbon number distribution of crude oil, and the results are shown in Figure 1.

![Figure 1. Composition of crude oil.](Image)

Natural gas and crude oil were fully stirred under the original formation conditions (pressure: 37 MPa and temperature: 89 °C) so that the preparation of live oil was consistent with the formation of crude oil. The preparation process was completed in a pressurize—volume—temperature (PVT) analysis system produced by the DBR Corporation of Canada. The basic parameters of the obtained live oil were tested, and the results are shown in Table 1.

2.1.2. Gas. The gas used for live oil preparation was obtained from formation of crude oil by flash separation. Table 1 shows the component of the gas obtained using an Agilent 6890 AGC gas chromatograph. Also, the gas used in the experiments sourced from CO₂ cylinders with a purity of 99.99%.

Table 1. Component of the Separated Gas

| Component | molar fraction (%) |
|-----------|-------------------|
| C₁        | 80.99             |
| C₂        | 7.94              |
| C₃        | 4.14              |
| C₄        | 1.33              |
| C₅        | 0.09              |
| N₂        | 5.06              |
| CO₂       | 0.18              |

2.1.3. Brine. To shield the signal of water and better display the signal of crude oil, deuterioxide (D₂O) was used to prepare simulated formation water in the experiment. Table 2 shows the ionic components of formation water that were determined using an IC761 ion chromatograph.

2.1.4. Core. All the cores used in the experiment were natural cores drilled from the formation in Ma 18 well area, Xinjiang. Also, these cores were low-permeability glutenite cores with an average gas permeability of only 0.16 mD and porosity of 8.84%. All the cores were standard cores with a diameter of 38 mm and a length of 70 mm.

2.2. Slim Tube Experiment. The slim tube method was usually adopted to measure the minimum miscibility pressure (MMP) of oil and CO₂ because the displacement process in the porous media provided by the slim-tube model could eliminate the influence of fluidity ratios, gravity differentiation, heterogeneity, and other factors to the greatest extent. Table 3 shows the parameters of the slim tube, filled with quartz sand.

The procedures for the slim tube experiment were as follows. First, the slim-tube model was put in an oven and heated to the experimental temperature (89 °C). The oil sample was saturated in the slim-tube model at an injection rate of 1 mL/min to make it reach the saturation state. Also, the crude oil was displaced with CO₂ at an injection rate of 0.1 mL/min, and the pore volume (PV) and oil recovery during CO₂ injection were recorded. After CO₂ injection with 1.2 times the PV, the gas injection was stopped.

2.3. Interaction between CO₂ and Crude Oil. The basic properties of crude oil and the effects of its interaction with CO₂ were measured using the DBR PVT analysis system. The equipment included a PVT cell, a thermostatic oven, a constant pressure pump, an altimeter, and an intermediate vessel. The experiment was carried out in the PVT cell that was a high-pressure and high-temperature visible glass cylinder with a visual volume of 280 mL. The maximum pressure and temperature were 70 MPa and 200 °C, respectively. Figure 2 shows a flow chart of the entire PVT device.

First, crude oil and natural gas samples were proportioned into the agitator according to gas-oil ratio (GOR) and reservoir temperature. Then, the oil and gas were stirred sufficiently to allow the gas to dissolve in the oil under conditions above the reservoir pressure. At constant temperature and pressure, the prepared live oil was transferred into the PVT cylinder.
Different mass percentages of CO_2 (0, 5, 10, 20, 30, 40, and 50%) were added into the PVT cylinder. Then, the gas was completely dissolved in the oil by pressurizing and stirring, and a single phase saturation state was reached. After the system reached a stable state, the effects of the injected gas on the saturation pressure, swelling factor, dissolved GOR, and viscosity of formation oil were analyzed.

2.4. CO_2 and Core Reaction Experiment. The low-permeability core was cut into several thin slices about 1 cm thick, as shown in Figure 3. The slices were then placed in a high-temperature and high-pressure corrosion-resistant reaction kettle, and sufficient amount of formation water was added. After sealing the reactor, a gas booster system was used to inject CO_2 gas into the reactor to 37 MPa, and the reactor was heated to formation temperature (89 °C) to allow adequate reaction between CO_2 and the core. A day later, we took one of the slices from the reactor and dried it in an oven. After drying, the mineral dissolution on the core surface was observed by SEM. Then, the core was ground into powder, and the changes in mineral composition and relative content of the core were analyzed using a X-ray diffractometer. The above steps were repeated to compare the effect of CO_2 on the core at different reaction times.

2.5. Nuclear Magnetic Resonance Experiment. The microscopic pore structure characteristics of low-permeability reservoir were studied by NMR detection technology. The CO_2 flooding experiment under the conditions of reservoir temperature and pressure was carried out to study the displacement mechanism of CO_2 on a microscale.

Schematic diagram of the displacement experimental device in Figure 4. First, the core was vacuumed and saturated with simulated formation water prepared with deuterioxide. The experimental temperature was set as reservoir temperature (89 °C), and the experimental pressure was set as reservoir pressure (37 MPa). Then, the oil sample was saturated in the core until the moisture content reached approximately 0% to establish the bound water and initial oil saturation. After oil saturation, the T_2 relaxation distribution and one-dimensional frequency coding of the core were tested.
and the grayscale and pseudo-color maps were generated at the same time. Finally, a CO₂ slug was then injected at a constant injection rate of 0.1 mL/min to displace the crude oil until the oil production became negligible. During the experiment, the $T_2$ relaxation distribution and one-dimensional frequency coding were collected continuously. Also, the oil and water distribution in the process of the experiment were imaging processing.

A low-field NMR device, MacroMR12-150H-I, which was produced by Newmai Analytical Instruments Co., Ltd., was used in this experiment. The core holder was customized for the NMR high-temperature and high-pressure core holder with a maximum compression capacity of 70 MPa. The principle of low-field NMR was to obtain the $T_2$ relaxation distribution of the sample through the echo attenuation signal collected from the sample after being saturated with water and the mathematical inversion calculation of the SIRT inversion algorithm. There were three relaxation mechanisms for fluids in rock pores, including volume relaxation ($T_{2b}$), surface relaxation ($T_{2s}$), and diffusion relaxation ($T_{2d}$), according to NMR fundamental principles, and the transverse relaxation time could be expressed using $\text{eq } 1.$\textsuperscript{24,25}

$$\frac{1}{T_2} = \frac{1}{T_{2b}} + \frac{1}{T_{2s}} + \frac{1}{T_{2d}}$$

(1)

In general, the lateral relaxation of hydrophilic rocks mainly depended on their surface relaxation and diffusion relaxation, and the volume relaxation was usually negligible. Moreover, when the echo interval and magnetic field gradient were small, the diffusion relaxation could also be ignored. $\text{Equation 2}$ is the transverse relaxation expression:\textsuperscript{17,19}

$$\frac{1}{T_2} \approx \frac{1}{T_{2s}} = \rho_s \left( \frac{S}{V} \right)_{\text{pore}}$$

(2)

where $\rho_s$ is the $T_2$ surface relaxation rate ($\mu$m/ms) and $(S/V)_{\text{pore}}$ is the specific area ($\mu$m$^2$/μm$^3$).

The $T_2$ relaxation distribution could be converted into the pore radius distribution according to certain conversion.\textsuperscript{26,27} Therefore, the $T_2$ distribution actually reflected the distribution of pore size.

### 3. RESULTS AND DISCUSSION

#### 3.1. Minimum Miscibility Pressure.

According to industry standards,\textsuperscript{28} when 1.2 times of PV gas is injected and its displacement efficiency is greater than 90%, it is considered that miscible state is reached at this time. Therefore, it is necessary to select two or more experimental points in both miscible and immiscible states. Then, two straight lines are fitted to the points in the two states, and the pressure at the intersection of the two lines is considered to be the MMP.

According to the actual formation conditions, five pressures of 25, 30, 32, 35, and 37 MPa were set for the slim tube tests. These points were divided into two groups, miscible and immiscible, for fitting. The experimental results are mapped in Figure 5. The intersection point of the two fitted lines was 33.6 MPa, which was termed the MMP.

#### 3.2. Physical Properties of Crude Oil.

In these experiments, the influence of CO₂ injection on the properties of crude oil was studied. Figure 6 shows the dependence of the pressure, swelling factor, GOR, and viscosity of crude oil on CO₂ injection volume.

![Flow chart of the flooding experiment.](image-url)

**Figure 4.** Flow chart of the flooding experiment.

![Minimum miscibility pressure of CO₂ and crude oil.](image-url)

**Figure 5.** Minimum miscibility pressure of CO₂ and crude oil.
Figure 6a shows the effect of injected gas volume on the saturation pressure. With the increase in the proportion of the CO₂ injection ratio, the pressure of the crude oil bubble point increased gradually. Due to the relatively large ratio of dissolved gas to oil in the formation and the limited capacity of dissolved gas, the saturation pressure increased greatly after gas injection. Figure 6b shows the relation of injected gas volume with the swelling factor. With the increase in the proportion of the CO₂ injection ratio, the swelling factor increased. Therefore, the volume of crude oil also increased. CO₂ injection was beneficial for solubilization and oil displacement. Figure 6c shows the relation curve between dissolved GOR and injected gas volume. The saturation pressure of crude oil also kept rising with the increase in CO₂ injection, and the dissolved GOR also gradually increased. Figure 6d shows the viscosity versus injected gas volume. CO₂ had a certain viscosity reduction effect on crude oil. Therefore, with the continuous CO₂ injection, the viscosity gradually decreased as the gas dissolved in the crude oil.

3.3. Dissolution Reaction. The original and aged core slices were taken out and dried. The slices were then compared under an environmental scanning electron microscope magnified 1000 times. As shown in Figure 7 the pores of the core were poorly developed and highly heterogeneous, and there were clay minerals distributed on the surface of the particles and inside the intergranular pores.

It could be seen from Figure 7 that the core surface had obvious dissolution phenomenon in an acidic environment. After 1 day of reaction, the mineral surface was found to become rough. After 2 days of reaction, the slices of cores were further dissolved, and clay particles and other deposits appeared on the mineral surface. These clay particles were the dissolution reaction products of minerals. After 3 days of reaction, a large number of clay particles were deposited on the mineral surface with some loose rocks.

Table 5 shows that the content of quartz changed little due to the stable physical and chemical properties of quartz, and it did not participate in the reaction between CO₂ and cores. However, CO₂ was dissolved in formation water to form carbonic acid, and H⁺, CO₃²⁻, and HCO₃⁻ were generated through ionization to form an acidic formation water environment. The reaction equations are shown in eqs 3–5.

\[
\text{CO}_2(g) \rightarrow \text{CO}_2(\text{aq})
\] (3)
With the increase in reaction time, the dissolution reaction of feldspar took place in an acidic environment, and clay minerals such as kaolinite are formed. Thus, feldspar content gradually decreased, while clay mineral content gradually increased. Equations 6–8 were the dissolution reactions of potassium feldspar, albite, and anorthite, respectively.

\[
\text{CO}_2(\text{aq}) + \text{H}_2\text{O} \rightarrow \text{H}_2\text{CO}_3 \quad (4)
\]

\[
\text{H}_2\text{CO}_3 \rightleftharpoons \text{H}^+ + \text{HCO}_3^- \rightleftharpoons 2\text{H}^+ + \text{CO}_3^{2-} \quad (5)
\]

As CO\textsubscript{2} was injected into the formation, it reacted with rocks and minerals in the formation. On one hand, the clay and other mineral particles generated after the dissolution reaction of rock minerals migrated and deposited with formation fluid and even blocked pores, causing damage to the reservoir. On

| time    | mineral content (%) | quartz | feldspar | clay |
|---------|---------------------|--------|----------|------|
| origin  | 35.0                | 42.3   | 22.7     |
| 1 day   | 35.1                | 36.9   | 27.9     |
| 2 day   | 36.9                | 33.4   | 29.8     |
| 3 day   | 37.2                | 31.6   | 31.4     |

Figure 7. Electron microscope scanning of cores with different reaction times.

Table 5. Core Mineral Composition at Different Reaction Times
the other hand, the reaction could improve the pore structure of the reservoir and increase the pore space. Simultaneously, under the action of dissolution, flow and scouring, particles gradually became smaller so as to realize the relief of plugging. This would improve seepage conditions and thus had a good effect on the development of LPHGRs.

3.4. Nuclear Magnetic Resonance. Figure 8 shows $T_2$ spectra at different moments of CO$_2$ displacement. Signal strength is proportional to the amount of oil in the core, and thus, the area difference between the two measurements at the $T_2$ relaxation distribution determines the recovery factor. The black line represents the distribution of initial oil saturation, and the other colors represent the saturation distribution at different injection times after CO$_2$ injection. The brown line represents the final state during 931 min of displacement. From the $T_2$ relaxation distribution, the change was quite great in the first 54 min, after which the remaining oil saturation did not decrease significantly.

According to the $T_2$ relaxation distribution, the core was divided into macropores ($T_2 > 40$ ms), medium pores (1 ms $< T_2 < 40$ ms), and small pores ($T_2 < 1$ ms). The distribution of crude oil in various pores accounted for 13.64, 44.63, and 41.73%, respectively. After a long period of CO$_2$ injection, no more oil was displaced, and the ultimate oil recovery rate was 35.98%, in which the recovery rates from macropores, medium pores, and small pores were 69.39, 46.07, and 14.27%, respectively. The experimental results showed that with the continuous injection of CO$_2$, the signal of the $T_2$ relaxation distribution was gradually weakened, which also indicated that crude oil was constantly displaced with gas. CO$_2$ mainly acted on the crude oil in the macropores and medium pores, while it had no obvious effect on crude oil in small pores.

Figure 8. $T_2$ spectra at different moments of CO$_2$ displacement.

Figure 9. Distribution of remaining oil at different positions during CO$_2$ displacement.
Figure 9 shows the distribution of remaining oil at different positions of cores with a total length of 50 mm in the process of CO2 flooding. The black line shows the distribution location of oil in the original oil-bearing state. The other three lines show the oil content of each part of the core at 60, 240, and 600 min, respectively. As known from the signal intensity of the 1-D distribution diagram, in the process of CO2 flooding, the remaining crude oil saturation water decreased with the elevated injection time.

Table 6 summarizes the NMR grayscale and pseudo-color images of core cross-sections at different times during CO2 displacement. The brighter areas in the image indicate a stronger hydrogen signal, which means that there were larger pores. For the sake of better observing the microscopic structure characteristics of rock samples, pseudo-color processing was carried out on the grayscale image to obtain the pseudo-color images of the oil phase distribution in pores at different moments in the process of CO2 displacement. In the pseudo-color image, the redder the color was, the stronger the hydrogen signal in that region was. The stronger the hydrogen signal was, the larger the pore throat of the rock sample was in this area, which was the dominant channel of the rock sample.

As clearly seen from the 1-D and 2-D images, the core had strong heterogeneity. After initiating CO2 injection, the pressure gradient was rapidly established, so the inlet oil volume decreased significantly. Meanwhile, a stable flood front was formed due to the miscibility of CO2 and crude oil. Therefore, the crude oil could be displaced from the macropores and medium pores.

4. CONCLUSIONS
In this paper, the displacement effect of CO2 in LPHGRs was researched. The main conclusions were drawn:

1. The MMP between CO2 and the crude oil was 33.6 MPa. Also with an increase of the proportion of CO2 injection, the saturation pressure, swelling factor, and GOR of crude oil increased, but the viscosity decreased.
2. The mineral composition of the rock was mainly quartz, feldspar, and clay minerals. SEM showed that the pores of the core were poorly developed and highly heterogeneous. The clay and other mineral particles produced by the dissolution reaction of rocks and minerals migrated and deposited with formation fluid to damage the reservoir. On the contrary, it also improved pore structure, increased pore space, and increased reservoir permeability.
3. a microscale, the miscible interaction between CO2 and crude oil formed a stable displacement front, which enabled crude oil to be well displaced from the macropores and medium pores but generated little effect on the small pores.

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