The phase behavior of reservoir fluids is crucial for ensuring the smooth operation of an underground gas storage (UGS). In this work, three experiments were conducted to investigate the phase behavior of condensate and crude oil systems within different development stages of gas storage in the Bohai Bay Basin of eastern China. In stage 1, constant volume depletion (CVD) test was conducted to simulate the interaction of the original condensate gas-live oil system. In stage 2, the phase behavior of a series of mixtures of live oil with 47.5, 58.3, 69.9, and 77.4 mol% of condensate gas was studied by constant composition expansion (CCE) test. In stage 3, a new experiment was designed to study the mass transfer and phase behavior during cyclic injection-production process of the underground gas storage. The results from the CVD test describe the effect of each pressure stage on produced gas composition and equilibrium oil properties in the condensate crude oil system. The results of the CCE test indicate that condensate gas can expand the oil and improve the properties. The results of cyclic injection-production experiment demonstrate that the mass transfer in condensate gas and oil system is a dual mechanism of condensation and vaporization. With the increase of the cycle, the intermediate components of oil phase are enriched and the heavy components decreases. After four cycles, the oil and gas interphase mass transfer reaches dynamic equilibrium. In addition, test results show that multicycle operation is beneficial to improve the oil properties. This work is of great significance to understand the phase transition mechanism of underground gas storage, which provides a basis for other researchers to further update and improve the EOS model.

1. Introduction

Natural gas with the advantage of high cleanliness and reliability has been regarded as an alternative in the low-carbon era [1–3]. The apparent consumption of natural gas has reached 3.22 × 10⁹ m³ by 2020 in China, which is still growing at a staggering rate of 8% per year [4, 5]. In addition, there are seasonal and regional imbalances in the supply and demand of natural gas in the Chinese market. For instance, due to winter heating supply in the north, natural gas consumption is three times much higher than in summer [6, 7]. Consequently, for the reasons of energy security and seasonal peak regulation, China is stepping up the construction of gas storage facility [8]. There are two feasible gas storage methods, namely, surface gas storage and underground gas storage (UGS). Among them, underground gas storage is popularized because of its larger storage capacity, higher security, and lower operational costs [9–11]. The method involves injecting gas into the underground porous medium in summer and withdrawing in winter [12, 13]. 25 underground gas storages have been put into operation in China, most of which are converted from depleted gas or oil reservoirs [14]. Knowledge of the phase behavior for formation fluid is the key to ensuring the stable and efficient production of the reservoir [15]. Complex phase equilibrium will occur when the gas is injected into the reservoir, such as tertiary oil recovery, CO₂ geological storage, and the gas injection.
process of the gas storage studied in this article [16, 17]. For a condensate reservoir, it is now agreed that cyclic gas injection to maintain pressure near or over the reservoir dew point pressure and prevent condensate banking or condensate losses [18–20]. Based on the CCE and CVD tests, Guo et al. [21] measured the dew point pressure and retrograded liquid volume of different rich condensate gas samples, respectively, and combined with a thermodynamic model to simulate the fluid phase behavior during the depletion development of this type of rich condensate gas. Abbasov and Fataliyev [22] built a specific laboratory equipment to investigate the volumetric properties, liquid drop-out conditions, dew point pressures, and fogging up of condensate gas during the cyclic dry gas injection. Subsequently, numerous studies have analyzed the effect of different injection parameters (such as gas category, full pressure maintenance, and partial pressure maintenance) on the phase state of condensate reservoir and have evaluated the effectiveness of cyclic gas injection [23–25]. Compared with conventional gas reservoir development, the operation of gas storage is quite different [26, 27]. Although they both have similar fluid systems and displacement methods, the development of oil and gas reservoirs is a directional, low-speed material exchange process. The operation of UGS has the characteristics of high injection-production intensity, large discharge volume, and cyclic injection-production [28, 29]. Generally, the operation process of gas storage needs completing gas injection (7–9 months) and gas production (3–5 months) in one year and start a new cycle in the second year [30, 31]. Therefore, the conventional reservoir development theories cannot be diametrically applied to the operation of the underground gas storage, which prompts many scholars to study the typical injection-production system of gas storage with high intensity, large displacement, and multiple cycles.

Based on laboratory experiments, many scholars have carried out mechanistic studies for the special injection-production characteristics of underground gas storage. As early as 1990s, Billiotte et al. designed the microdisplacement experiment to investigate the effect of wettability and capillary force on the cyclic injection-production process of gas storage. The results show that when the number of nonwetting cycles generated during injection increases, the gas recovery will be significantly reduced in withdrawal process [32]. At the same time, some literatures have also clarified the mechanism of water invasion and gas channeling during the process of cycle injection-production in gas storage [28, 33, 34]. Zeng [35] confirmed the existence of a significant relative permeability hysteresis during cyclic injection-production in gas storage using indoor experiments. During the cyclic injection-production process, the permeability of the nonwetting phase decreases, and the copermeability zone of the two phases shrinks, which directly affects the injection-production ability and the capacity of the UGS [35]. Wang et al. [36] took cores in the actual gas storage and conducted experiments to simulate the initial injection and subsequent multiple injection-production process, exploring the intrinsic relationships between reservoir capacity and pressure, and between reservoir capacity and injection and production rounds. Shao et al. [37] studied the periodic changes in rock pore structure due to cyclic injection-production of underground gas storages. Their experimental results concluded that physical properties of rocks are severely damaged by alternating stress, which can substantially enhance the condensate recovery while completing the operation of the gas storage.

The previous work has mainly focused on mechanisms of multiphase flow percolation during the injection-production process. However, for an underground gas storage converted from a condensate gas reservoir, the phase state of the reservoir fluid in porous medium during the cyclic injection-production is dynamic and complex [38]. Recently, Tang et al. [39] reported the phase behavior of condensate gas and injected dry gas during cyclic injection-production in a condensate reservoir-type gas storage. The cyclic gas injection process is considered conducive to the evaporation of retrograde fluid, which can substantially enhance the condensate recovery while completing the operation of the gas storage.

As found in the literature, the current stage of indoor experimental studies of underground gas storage are heavily weighted on the seepage characteristics of cyclic injection,
and the characteristics of gas phase changes after cyclic gas injection in condensate reservoir-type gas storage. At present, there is not a complete consensus on how the phase equilibrium changes of crude oil and condensate gas system in gas storage system. Meanwhile, according to our literature research, the existing experimental method on the phase state of condensate gas and crude oil is not applicable to gas storage system, especially the multicycle injection-production process. The objective of this study is (1) understand the phase behavior of condensate gas and crude oil systems during its depletion development stage and gas storage operation stage and (2) explain the mechanisms underlying these change processes. This outcome should assist in understanding the phase transition mechanism of underground gas storage in the full life cycle.

2. Workflow

This part involves the workflow of experimental method to investigate the phase behaviors of condensate gas and crude oil system in the underground gas storage. In this work, taking an example of a complex condensate reservoir-type gas storage in the Bohai Bay Basin of eastern China, the investigated steps include the following: (1) Based on the CVD experiment, the interaction of the live oil-condensate gas system is simulated during the depletion development stage, that is, the stage before the construction of the gas storage. (2) After the CVD experiment, the phase behavior of the mixture of live oil and condensate gas at different proportions was investigated using the CCE experiment, which corresponds to the initial stage of gas storage construction. (3) The new designed cyclic injection-production experiment was used to simulate the gas storage stage. The main objective is to better understand the mass transfer and phase behavior for condensate gas and live oil under truly reservoir conditions, during the cyclic injection-production process. Figure 1 displays the workflow of the investigation method.

3. Experiments

3.1. Materials. Fluid samples must be taken in the early stage of reservoir development to obtain samples truly representative of the reservoir fluid [40]. The crude oil sample and condensate gas sample were collected from gas storage in eastern China. Because exploitation has been undertaken for more than decades before being converted into gas storage, the original formation fluid samples are currently

| Table 1: Composition of dead oil sample. |
| Component | mol % | wt % |
| iC4 | 0.05 | 0.02 |
| nC4 | 0.11 | 0.03 |
| iC5 | 0.30 | 0.12 |
| nC5 | 0.56 | 0.21 |
| C6 | 5.28 | 2.41 |
| C7 | 17.68 | 9.01 |
| C8 | 13.07 | 7.42 |
| C9 | 4.33 | 2.78 |
| C10 | 4.14 | 2.94 |
| C11 | 4.13 | 3.22 |
| C12 | 3.56 | 3.04 |
| C13 | 4.06 | 3.77 |
| C14 | 4.28 | 4.31 |
| C15 | 3.73 | 4.08 |
| C16 | 2.95 | 3.47 |
| C17 | 2.81 | 3.54 |
| C18 | 2.38 | 3.17 |
| C19 | 3.98 | 5.55 |

| Component | mol % | wt % |
| C20 | 3.47 | 5.06 |
| C21 | 2.29 | 3.50 |
| C22 | 2.26 | 3.63 |
| C23 | 2.27 | 3.82 |
| C24 | 1.94 | 3.42 |
| C25 | 1.96 | 3.57 |
| C26 | 1.54 | 2.94 |
| C27 | 1.45 | 2.87 |
| C28 | 1.14 | 2.33 |
| C29 | 1.10 | 2.33 |
| C30 | 0.74 | 1.62 |
| C31 | 0.76 | 1.72 |
| C32 | 0.65 | 1.52 |
| C33 | 0.46 | 1.12 |
| C34 | 0.27 | 0.68 |
| C35 | 0.18 | 0.46 |
| C36 | 0.12 | 0.32 |
| C37 | 0.14 | 0.32 |

| Table 2: Composition of the solution gas. |
| Component | mol % | wt % |
| C1 | 68.75 | 46.61 |
| C2 | 10.54 | 13.40 |
| iC4 | 0.93 | 2.29 |
| nC4 | 10.84 | 26.64 |
| CO2 | 2.68 | 3.63 |
| N2 | 6.26 | 7.43 |

| Component | mol % | wt % |
| C1 | 68.75 | 46.61 |
| C2 | 10.54 | 13.40 |
| iC4 | 0.93 | 2.29 |
| nC4 | 10.84 | 26.64 |
| CO2 | 2.68 | 3.63 |
| N2 | 6.26 | 7.43 |

| Table 3: Measured live oil properties at 25.02 MPa and 89°C. |
| Live oil properties |
| Solution GOR (sm³/m³) | 213 |
| Density (g/cm³) | 0.6251 |
| Viscosity (mPa·s) | 0.246 |
| Bubble point pressure (MPa) | 25.02 |
| Crude oil volume factor | 1.68 |

| Component | mol % | wt % |
| N2 | 0.13 | 0.14 |
| CO2 | 0.85 | 1.49 |
| C1 | 77.61 | 49.48 |
| C2 | 10.60 | 12.67 |
| C3 | 5.14 | 9.01 |
| iC4 | 0.23 | 0.66 |
| nC4 | 0.57 | 1.63 |
| C6 | 0.48 | 1.60 |
| C7 | 1.43 | 5.46 |
| C8 | 0.56 | 2.38 |
| C9 | 0.72 | 3.46 |
| C10 | 0.47 | 2.50 |
| C11 | 1.21 | 9.52 |

| Molecular weight of C11+ | 198.7 |
| Relative density of C11+ | 0.822 |
| Density of stock tank oil (g/cm³) | 0.787 |
| Gas-oil ratio (sm³/m³) | 2587 |
| Dew point pressure (MPa) | 24.76 |

| Component | mol % | wt % |
| N2 | 0.13 | 0.14 |
| CO2 | 0.85 | 1.49 |
| C1 | 77.61 | 49.48 |
| C2 | 10.60 | 12.67 |
| C3 | 5.14 | 9.01 |
| iC4 | 0.23 | 0.66 |
| nC4 | 0.57 | 1.63 |
| C6 | 0.48 | 1.60 |
| C7 | 1.43 | 5.46 |
| C8 | 0.56 | 2.38 |
| C9 | 0.72 | 3.46 |
| C10 | 0.47 | 2.50 |
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| Dew point pressure (MPa) | 24.76 |
unavailable. Therefore, the characteristics of the original crude oil and condensate gas were recovered by the gas-oil ratio and saturation pressure method, respectively, in this study [41]. Upon completion of recombining, fluid samples are examined to ensure they are representative of the original formation fluid properties.

3.1.1. Oil Sample. The provided dead oil sample was cleaned and filtered to remove any fine brine and solid impurities. The composition was also analyzed using an Agilent 7890B gas chromatograph; the results are shown in Table 1. The recombined live oil was prepared by mixing the solution gas and dead oil according to the gas-oil ratio at 26 MPa and 89°C (the original reservoir condition). The solution gas was supplied by Beijing Yong sheng Gas Tech Co., Ltd., and the specific composition of the solution gas is as described in Table 2. The bubble-point pressure, solution gas-oil ratio, density, viscosity, and volume factor of the live oil sample are listed in Table 3. These data indicate that the oil is characterized as near-critical oil, also called volatile oil, which contains a high content of light to intermediate hydrocarbons.

3.1.2. Condensate Gas Sample. Condensate gas was sampled using the separator sampling method. The separator oil sample was chemical analyzed with an Agilent 7890B chromatograph. The in situ condensate gas was obtained by recombining the separator oil and the natural gas accord to the dew point pressure. The composition of condensate gas, C_{11+}, molecular weight, C_{11+} relative density, stock tank oil density, gas-oil ratio, and dew point pressure are listed in Table 4.

3.2. Apparatus. Phase behavior tests for condensate gas-crude oil system are conducted on a PVT apparatus. Figure 2 depicts the schematic diagram of the experimental laboratory system. The high-pressure system consisted of a PVT container, constant temperature air bath, sample cylinder, vacuum pump, syringe pump, gas meter, gas chromatograph, rolling ball viscometer, densitometer, and operation and recording system. The PVT container can withstand the maximum working pressure and temperature of 70 MPa and 180°C. The real-time pressure was monitored by the built-in manometer with an accuracy of 0.01 MPa. The desired experimental temperature is controlled with ±0.1°C by a heating jacket wrapped around the PVT cell. The total internal volume of PVT cell is 350 cm³, and the dead volume is 1.41 cm³. The maximum uncertainty in volume measurements is ±0.01 cm³. The real-time volume, pressure, and temperature data were recorded and shown on a display panel. The test fluids were precisely introduced cell by a syringe pump (100DX, Teledyne ISCO) at a constant pressure during the experiment. A magnetic stirrer is mounted at the top of the PVT cell to accelerate equilibrium. The test fluid cylinders are maintained in an air-bath oven at the experimental temperature. Oil viscosity was measured by rolling ball viscometer, and oil density was determined using a density meter (SVM3000, Anton Par). The accuracy in viscosity and density is estimated less as 1% and ±10^-4 g/cm³, respectively.

3.3. Experimental Procedures

3.3.1. Constant Volume Depletion (CVD) Test. The CVD test is assuming the immobility of the retrograde fluid of a rock pores to simulate an actual reservoir depletion [42]. The procedure in this experimental section is slightly different from that of conventional CVD test. Specifically speaking, the fluid injected into the PVT cell is no longer a single condensate gas phase but is replaced by the condensate gas-crude oil mixture. This is more in line with the real development process of this type of condensate gas reservoir with oil ring.

Firstly, experimental preparations are performed before the start of the formal tests, including cleaning, leakage testing,
volume calibration, vacuuming, and sample generation. The PVT cell and tubes are cleaned by petroleum ether and ethanol in sequence; then, high-pressure nitrogen is injected into the PVT cell to perform the leakage testing. After that, water is injected to calibrate the efficient volume. Next, the whole system is douched and dried by the injected air and vacuumed at 1 kPa. At the last step, the recombined samples (condensate gas, live oil) were transferred to the cylinder from the formation fluid sampling equipment and the air bath oven is set as the reservoir temperature (89°C) for 24 hrs.

The exact volume of condensate gas and live oil at injection pressure and temperature had been calculated in advance [43]. Condensate gas and live oil were precisely transmitted into the PVT cell sequentially, and the temperature was maintained at the test temperature by using the heating jacket. Then, the fluid in the PVT cell was pressed and held to a level approximately 4 MPa than the initial reservoir pressure and the magnetic stirrer kept stirring. The equilibrium process sustained for at least 24 hrs. Thereafter, the pressure decreased to the initial reservoir pressure.
At each step, the stirrer was turned on for 0.5 hours to stimulate the mass transfer between condensate gas phase and live oil phase. Then, we maintain the pressure for 2 hours to reach a new gas-liquid equilibrium. Thereafter, the vent valve at the top of PVT cell was slowly opened to release the gas until the volume is restored to the initial constant volume. During this process, the pressure in the PVT cell was held at the current level through the piston and the vent gas was sampled to analyze composition by the gas chromatographic. The viscosity and density of equilibrium oil at each step were also measured at each pressure step. Finally, the pressure declined to the desired low point ($P_d = 4$ MPa), and chromatographic analysis of the residual oil phase was carried out.

3.3.2. Constant Composition Expansion (CCE) Test. The constant composition expansion (CCE) test is a common fluid phase experiment, and the oil composition in the PVT cell is kept constantly while the volume is expanded as the pressure is gradually depleted. The CCE test is usually used to determine the two-phase volumetric behavior of oil-gas mixture under bubble-point pressure, such as relative volume and isothermal compressibility [44].

In this section, four CCE experiments of the live oil and condensate gas mixtures (47.5, 58.3, 69.9, and 77.4 mol% of condensate gas) were performed using the PVT apparatus. The oil sample is obtained from the equilibrium oil at the last pressure step of the CVD experiment in the previous section, that is, the formation oil under reservoir depletion pressure before the construction of the gas storage. Firstly, the calculated volume of condensate gas and live oil were introduced at 89°C. The pressure was increased using the high-pressure pump at the bottom of the PVT cell. Thereafter, the magnetic stirrer started to work until the gas completely dissolved and a single liquid phase is formed. Depressurization steps were started at 40 MPa, far from the bubble-point pressure. At each depressurization step, the fluid is stirred constantly to promote gas release. After that, a minimal equilibrium time of 30 minutes was guaranteed. Finally, the volume of the mixture was recorded at each pressure step when the volume and pressure variation were less than 0.03 cm$^3$ and 0.03 MPa.

3.3.3. Cyclic Injection-Production Experiment. The well-designed cyclic injection-production experiment of the condensate gas and crude oil took place in the PVT cell. It would enable to accurately simulate the phase behavior of condensate gas and crude oil in the gas storage. This experiment was carried out immediately after the last pressure step of the CVD test.
Firstly, a certain amount of condensate gas was introduced into the PVT cell until the pressure reached 26 MPa ($P_{up}$), which is the upper limit pressure of the gas storage. In this process, the volume of oil and gas mixture in the PVT cell was kept constant. Additionally, the fluids were stirred constantly and kept at equilibrium at least for 2 hrs. Thereafter, we took a spot of the equilibrium oil sample at the bottom valve for the composition, gas-oil ratio, density, and viscosity measurements. After that, the vent valve in the top was opened to release the gas until the system pressure dropped to 8 MPa ($P_{low}$), which was the lower limit pressure of gas storage. During this process, the volume of PVT cell is kept constant. Moreover, we sampled the produced gas and measured the composition. Finally, after the oil phase reached to a new equilibrium at the lower limit pressure, the physical properties and composition of the equilibrium oil were measured, respectively. This similar injection-production cycle was carried out 6 times. At each cycle, the properties and composition of the equilibrium oil were tested, as well as the produced gas. The whole cyclic procedure is depicted in Figure 3.
4. Results and Discussions

4.1. Constant Volume Depletion (CVD) Test. The CVD test is used to simulate the gas reservoir depletion development process before the construction of the gas storage. At each step of a CVD test, the pressure of condensate gas-oil system was declined and the gas was allowed to be released. Table 5 illustrates the data of separator gas during depletion process. The results show that the composition changes are not completely monotonous. Methane shows a trend of first increasing and then decreasing. However, the variation of the corresponding intermediate components (C₃ ~ C₈) is in the opposite direction. Moreover, the heavy component (C₉+) significantly reduced. These experimental results can be explained by a condensate gas retrograde behavior. In the early pressure reduction process, due to the condensate drop out, the intermediate and heavy components in the separator gas were reduced. After that, when the pressure is below the maximum condensation pressure, condensate retrograde gradually weakened and the content of intermediate components rise again. In addition, the fraction of ethane in the gas phase also increases, presumably due to the liberation of light components dissolved in crude oil during the depressurization process.

At each step, the equilibrium oil properties were immediately measured after the pressure stabilizes. It is found out that pressure depletion has a momentous effect on the fluid properties, such as GOR, density, and viscosity data. We take density and viscosity as examples, as shown in Figure 4. As pressure decreases, both density and viscosity of equilibrium oil progressive increase. The results are considered to be caused by the release of dissolved gas during depressurization process which also can be confirmed by the GOR variation curve in Figure 5, when the pressure depleted from 26 MPa to 4 MPa, the gas oil ratio decreases from 245.6 m³/m³ to 29.7 m³/m³. Figure 6 depicts the composition of the equilibrium oil at the last pressure step (depleted pressure) and compared with the original crude oil. The general trends of two oil samples are similar and the mole fraction of C₇₊ declines by increasing the carbon number. Additionally, the composition of methane of oil sample at the depleted pressure is well below that of the original oil and C₇ has the highest concentration in the former. After each depressurization step, retrogradation of gas and release of dissolved gas occur simultaneously. Intermediate components in the gas phase condense into and enrich the oil phase. But limited by the volume of retrograded liquid, this improvement of oil properties is marginal. On the contrary, a large amount of dissolved gas is released from the saturated live oil during depressurization, and as a consequence, the oil samples become heavier.

4.2. Constant Composition Expansion (CCE) Test. The crude oil and condensate gas were mixed with different gas concentrations: 47.5%, 58.3%, 69.9%, and 77.4% to conduct CCE test. The relative volume \( V_{rel} \), meaning the rate between the total volume of PVT cell of each stage at current pressure and the total volume at bubble-point pressure, is defined as follows:

\[
V_{rel} = \frac{V_{(T,P)}}{V_{(T,P_b)}}.
\]

In Figure 7, the pressure versus relative volume of crude oil and condensate gas system at the reservoir temperature are depicted. From these curves, CCE test was performed on all four concentrations, and experimental data has a smooth decline. But the P-V curve becomes continuous.
and inflection point is no longer obvious when the injected concentration increases.

Bubble-point pressure was identified by two different methods, as follows: (i) the inflection point of the slope for pressure-volume line and (ii) the first bubble was observed by the camera located in front of sapphire visual windows [45]. The first bubble escapes from the oil when the pressure was below the bubble-point pressure, and then, a large amount of gas converges a separate gas phase at the top PVT cell as the pressure decreased.

Swelling factor (SF) is defined as the rate between of the total molar volume \( V_{od(T,P_b)} \) of dissolved gas and oil at the bubble-point pressure \( P_b \) and reservoir temperature \( T \) and the molar volume of oil \( V_{o(T,P_o)} \), without the injectant) at the original pressure \( P_o \) and temperature [46]:

\[
SF = \frac{V_{od(T,P_b)}}{V_{o(T,P_o)}} \times \frac{1}{1 - x}.
\]

The measured bubble-point pressure \( P_b \) and swelling factor \( SF \) of each injected concentration are presented in Figure 8. The results show that \( P_b \) and \( SF \) increase with increasing injected concentration. The results illustrate that when the system pressure increases, the mixture with lower concentration of the injected is faster in reaching the bubble-point pressure faster and forms a single-phase fluid.

For example, \( P_b \) is nearly doubled when the injected concentration increases from 47.5% to 77.4 mol%. Meanwhile, SF also increases from 1.331 to 2.265 at the same time, which reveals that live crude oil at reservoir depleted pressure has excellent expansion property. Overall, the rate of increase of \( P_b \) and SF is faster when the injected concentration exceeds 47.5 mol%.

Oil density was also obtained using mass balance calculation. Because the mass of the mixture is conserved in the experimental process, the oil density at different experimental conditions can be calculated by

\[
\rho_{d(T,P)} = \frac{m_o(T,P) + m_{g(T,P)}}{V_{m(T,P)}}.
\]

Here, \( m_o(T,P) \) is the mass of live oil, \( m_{g(T,P)} \) is the mass of condensate gas at each concentration, \( V_{m(T,P)} \) is the total

| Component | Injectant | Composition of produced gas (mol %) |
|-----------|-----------|---------------------------------|
|           | 1         | 2     | 3     | 4     | 5     | 6     |
| N\textsubscript{2} | 0.13 | 0.66 | 0.28 | 0.20 | 0.17 | 0.15 | 0.14 |
| CO\textsubscript{2} | 0.85 | 1.04 | 0.93 | 0.88 | 0.87 | 0.86 | 0.86 |
| C\textsubscript{1} | 77.61 | 81.74 | 80.91 | 80.39 | 80.14 | 79.98 | 80.07 |
| C\textsubscript{2} | 10.60 | 9.22 | 9.93 | 10.24 | 10.33 | 10.43 | 10.41 |
| C\textsubscript{3} | 5.14 | 3.22 | 4.07 | 4.49 | 4.63 | 4.75 | 4.78 |
| iC\textsubscript{4} | 0.00 | 0.13 | 0.08 | 0.06 | 0.04 | 0.02 | 0.01 |
| nC\textsubscript{4} | 0.00 | 1.14 | 0.69 | 0.46 | 0.35 | 0.23 | 0.14 |
| iC\textsubscript{5} | 0.23 | 0.11 | 0.14 | 0.16 | 0.17 | 0.18 | 0.18 |
| nC\textsubscript{5} | 0.57 | 0.24 | 0.33 | 0.39 | 0.43 | 0.44 | 0.45 |
| C\textsubscript{6} | 0.48 | 0.37 | 0.38 | 0.37 | 0.38 | 0.38 | 0.38 |
| C\textsubscript{7} | 1.43 | 0.83 | 0.87 | 0.89 | 0.91 | 0.93 | 0.94 |
| C\textsubscript{8} | 0.56 | 0.41 | 0.38 | 0.37 | 0.36 | 0.36 |
| C\textsubscript{9} | 0.72 | 0.17 | 0.22 | 0.25 | 0.28 | 0.30 | 0.31 |
| C\textsubscript{10} | 0.47 | 0.14 | 0.15 | 0.16 | 0.17 | 0.17 | 0.18 |
| C\textsubscript{11+} | 1.21 | 0.58 | 0.64 | 0.69 | 0.76 | 0.82 | 0.79 |
volume of the mixture at each pressure step. All data were measured while the mixtures were above the bubble point pressure (single-phase fluid in the PVT cell). The calculated densities of the crude oil with four different gas concentrations are plotted as a function of pressure in Figure 9. As expected, density decreases with increasing injected molecular concentration under a constant system pressure and increases with increasing pressure under a constant gas molecular concentration.

4.3. Cyclic Injection-Production Experiment. In the previous sections, we tested the (1) phase behavior of the oil and gas system during constant volume depletion and (2) phase behavior of the crude live oil and condensate gas system at different gas injection ratios. To understand how the gas interacts with oil in gas storage, the cyclic injection-production experiment was performed.

One of the most important characteristics of gas storage is the composition of produced gas, which is related to the construction of the ground separation device and the control of the gas supply cost. Table 6 summarizes the measured composition of produced gas for each production cycle conducted at 20 MPa and 89°C. Equilibrium gas compositions were calculated by combining the measured separation gas and condensate compositions. In the first gas production cycle, the GC analysis data show increase in methane fraction and progressive reduction of C11+ in comparison with injected gas composition. Moreover, ethane and propane in produced gas tend to have different magnitudes of decrease. The above results indicate that the gas retrograde and heavy components drop out during the depressurization process. It is interesting to remember that nitrogen and butane are almost not contained in the injected gas, but the above two components were detected in a gas sample of the first production cycle. This means that there is not only gas condensation between injected gas and volatile oil but also some light components in the volatile oil also vaporize and are extracted along with the produced gas. Combining the results of the six gas production cycles, it can be found that the methane fraction in the produced gas shows a decreasing trend as the cycle increases, until it reaches equilibrium in the last two cycles. In contrast, the intermediate component (C2 - C10, except C4) and heavy component (C11+) in gas phase increases from the 1st cycle to 5th cycle and stabilize in the last two cycles. By comparing results, the composition of produced gas becomes closer to that of injected gas as the cycle further. Gas readily

| Component | Composition of equilibrium oil (mol %) |
|-----------|--------------------------------------|
| N2        | 0.19 0.09 0.07 0.06 0.07 0.07       |
| CO2       | 0.91 0.72 0.71 0.71 0.72 0.72       |
| C1        | 53.66 54.53 55.12 56.13 56.72 56.86 |
| C2        | 10.60 10.81 10.94 10.98 11.02 11.04 |
| C3        | 6.03 6.52 6.73 6.75 6.77 6.77       |
| iC4       | 0.11 0.07 0.03 0.02 0.01 0.00       |
| nC4       | 1.21 0.54 0.21 0.08 0.02 0.01       |
| iC5       | 0.38 0.43 0.45 0.46 0.45 0.45       |
| nC5       | 0.94 1.11 1.19 1.21 1.23 1.17       |
| C6        | 1.32 1.31 1.27 1.24 1.22 1.21       |
| C7        | 5.21 5.01 4.97 4.88 4.79 4.76       |
| C8        | 2.77 2.66 2.44 2.30 2.20 2.16       |
| C9        | 2.29 2.57 2.84 2.97 3.01 3.08       |
| C10       | 2.24 2.41 2.47 2.51 2.54 2.57       |
| C11+      | 12.14 11.22 10.56 9.70 9.23 9.13     |

Table 7: Composition change of equilibrium oil at upper limit pressure during the cyclic injection-production process.

| Component | Composition of equilibrium oil (mol %) |
|-----------|--------------------------------------|
| N2        | 0.05 0.03 0.02 0.02 0.02 0.02       |
| CO2       | 0.49 0.46 0.45 0.44 0.45 0.44       |
| C1        | 21.17 21.44 21.68 21.82 21.93 21.99 |
| C2        | 8.26 8.43 8.51 8.56 8.57 8.48       |
| C3        | 7.12 7.73 7.86 7.95 7.91 7.87       |
| iC4       | 0.22 0.11 0.05 0.03 0.00 0.00       |
| nC4       | 2.33 1.12 0.41 0.13 0.04 0.01       |
| iC5       | 0.73 0.82 0.88 0.91 0.92 0.93       |
| nC5       | 1.94 2.18 2.40 2.47 2.53 2.55       |
| C6        | 29.5 2.94 2.99 3.00 3.04 3.07       |
| C7        | 11.47 11.94 12.24 12.38 12.46 12.55 |
| C8        | 6.57 6.34 6.09 5.94 5.91 5.92       |
| C9        | 5.46 6.13 7.19 7.68 8.15 8.45       |
| C10       | 4.44 4.71 4.87 4.96 5.04 5.11       |
| C11+      | 26.80 25.62 24.36 23.71 23.03 22.61 |

Table 8: Composition change of equilibrium oil at lower limit pressure during cyclic injection-production process.

![Figure 10: Original oil and oil samples at the end of six cycles.](http://pubs.geoscienceworld.org/gsa/lithosphere/article-pdf/doi/10.2113/2022/4366803/55217814366803.pdf)
condensates and drops out in crude oil in the first cycle; however, when the oil phase is enriched with intermediate and heavy component, gas already starts to lose its condensation power in cycle 5. On the other hand, we have noticed that nitrogen and butane fraction also gradually decrease. This is due to the decrease in the content of the remaining nitrogen and butane components in the crude oil as the injection-production cycle increases.

Figure 10 displays the original oil and oil samples at the end of six cycles. The appearance of all oil samples are dark brown, and it is hard to differentiate by the naked eye. Therefore, the composition was analyzed by using GC–MS, and the equilibrium oil phase composition was recombined according to the separated gas oil ratio. Tables 7 and 8 list the composition of the crude oil at the upper and lower limit pressure in each cycle, respectively. The general trend of oil composition at the upper and lower limit pressures is similar. When the cycle increases, both the composition of methane in the two equilibrium oil phases increases while the heavy component fraction shows a decreasing trend. The intermediate component changes are influenced by the composition of the injected gas, and there is no obvious consistent pattern. The heavy component significantly decreases.

The composition data shows that complex mass transfer phenomena exist between the injected gas and crude oil in the cyclic injection-production of gas storage. The condensation in the gas phase and the vaporization in the oil phase take place simultaneously. In the first 4 cycles, condensation is significantly stronger than vaporization, resulting in an increase in intermediate components in the equilibrium oil.
phase. However, when the equilibrium oil phase is enriched and gradually approaches the injected gas component, the intermediate component transfer from gas to liquid composition also becomes less intense with cycle increase. Accompanying the increase in the number of contacts, the mass transfer between gas and oil reaches a dynamic balance, and oil composition tends to be stable. That is, oil and injected gas are in equilibrium in the PVT cell, and vaporization and condensation occur at identical intensities.

At each cycle, oil samples were taken to measure gas oil ratio density and viscosity. The equilibrium oil properties at each of cycle are presented in Figures 11 and 12. The number “0” cycle represents the oil phase before experiment, that is, crude oil at the pressure before gas storage construction. The decimal number (such as 0.5) and integer number (such as 1) corresponds to the upper limit and lower limit pressures. Firstly, in the building stage, the gas-oil ratio climbs rapidly along with gas injection, and at the upper reservoir pressure, the gas-oil ratio reaches 325 m³/m³, as shown in Figure 11. In the subsequent withdrawal stage, the saturated oil is degassed, and the gas-oil ratio decreases sharply to 72.2 m³/m³ at the lower limit pressure. The variation in density and viscosity correspond to the dissolution and release of the gas. Overall, the gas oil ratio of both equilibrium oils shows the same increasing trend with the increasing cycle. This is because of the condensation of the injected gas during cyclic injection-production. The enrichment of the intermediate components and the reduction of the heavy components in the equilibrium oil are conducive to the gas dissolution. However, due to the decrease of condensate intensity, the increase of gas-oil ratio slows down when the cycle increases. Correspondingly, the density and viscosity measurement results are shown in Figure 12. Due to the enrichment of the light and intermediate components in the equilibrium oil phase, the density and viscosity at the upper and lower pressures decline with the increasing cycle. Additionally, after 5 operation cycles, the density and viscosity gradually stabilize when the oil and gas transfer reach equilibrium.

5. Summary and Conclusions

In this work, a series of experiments were designed and employed to simulate the development and operation process for underground gas storage. Based on the experimental results discussed in this paper, the main conclusions can be drawn:

(1) The CVD test indicates that the composition of the produced gas at each pressure level shows a nonmonotonic variation depending on the intensity of the condensate retrograde. In addition, a decrease in pressure will increase the density, viscosity, and heavy component content of the oil phase.

(2) In the CCE test, the bubble-point pressure of oil is nearly doubled by increasing the condensate gas concentration from 47.5% to 77.4 mol% and the swell factor also increases from 1.331 to 2.265 at the same time. The high solubility of condensate gas can expand crude oil at reservoir condition.

(3) The results of cyclic injection-production experiment demonstrate that the mass transfer in condensate gas and crude oil system is a dual mechanism of condensation and vaporization. With the increase of the cycle, the content of intermediate and heavy components in the produced gas increases. After 4 cycles, the composition of the produced gas is close to that of the injected gas.

(4) The general trend of oil composition at the upper and lower limit pressures is similar. In the first 4 cycles, condensation is significantly stronger than vaporization, resulting in an increase in intermediate components in the equilibrium oil phase. After that, the mass transfer between two phases of gas and oil reaches a dynamic balance. Meanwhile, the density and viscosity of equilibrium oil are reduced, and the gas oil ratio increases. Multicycle operation is beneficial to improve the properties of the oil.

Data Availability

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

Conflicts of Interest

The authors declare no conflicts of interest.

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