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Impacts of CO_2-CH_4 Mixed Gas on Property of Formation Oil from the Bohai Oilfield

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Abstract: Mechanism analysis and technical scheme optimization on CO_2 displacement and CO_2 storage are based on the high-pressure physical properties of CO_2-added formation oil. Oil and natural gas samples from the BZ25-1 block in the Bohai oilfield were used to conduct high-pressure physical property experiments to explore the impacts of CO_2-CH_4 mixed gas on the properties of formation oil. After injecting different amounts of mixed gas, the saturated pressure was measured by constant mass expansion test, the viscosity was measured by falling ball method, the expansion coefficient was measured by gas injection expansion test, and the gas–oil ratio and volume coefficient were obtained by single degassing test. The results show that with gas injection, the saturation pressure and dissolved gas–oil ratio of formation oil increase, the volume coefficient and expansion factor go up, while the oil viscosity reduces. With the increase in gas addition, the properties of formation oil continue to improve, but the increase in improvement becomes flat. With the increase in pressure, the amount of dissolved gas in the formation oil will also increase. High-purity CO_2 is more helpful to change the properties of formation oil, while the gas mixed with CH_4 is more beneficial to elevate the formation energy. For the BZ 25-1 block, the gas injection amount of about 80 mol% is appropriate and the CO_2 purity of 60% can well balance the oil properties improvement and the formation pressure elevation.

Keywords: oil flooding by CO_2; CH_4 reinjection; mixed gas; property of formation oil; experimental study

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1. Introduction

Oil flooding by CO_2 injection can improve the effect of crude oil exploitation and realize geological CO_2 storage. CO_2 flooding and storage technology is one of the important ways to achieve the goal of "carbon peak and carbon neutral" [1,2], which has good economic and social benefits. CO_2 EOR technologies have received increasing emphasis [3,4]. Offshore oil reservoirs are rich in oil and gas reserves, but the degree of production is low. On the one hand, compared with onshore oil reservoirs, offshore oil reservoirs have high development costs, relatively low productivity, and a poor development economy. On the other hand, in order to expand economic benefits, after CO_2 flooding, the associated gas produced by oil wells is mixed with CO_2 and then reinjected. In this case, the relationship between gas concentration and fluid properties can be effectively grasped through the experiment of injecting mixed gas at high pressure, which can provide reference opinions for oilfield development. The Bohai Bay Basin has large geological reserves, and the potential reserves suitable for CO_2 flooding are up to 1.7 billion tons, which is the most important target of CO_2 flooding. By optimizing the design of the CO_2 flooding scheme, the reserves of low permeability reservoir in the Bohai Sea can be developed efficiently.

The impacts of CO_2 injected into the formation oil are the basis for analyzing CO_2 flooding and storage mechanisms and optimizing the scheme of CO_2 flooding.

The associated gas produced by oil production wells is mainly CH_4 [5,6]. For offshore fields, it is expensive to transport the associated produced gas onshore for decarbonization. From the perspective of cost, when CO_2 flooding is conducted in offshore oil fields, the associated gas produced by oil wells is mostly handled by local reinjection after mixing with CO_2. Therefore, when optimizing the design of the CO_2 flooding scheme in offshore fields, it is necessary to understand the impacts of CO_2 addition and purity (mixed with CH_4) on the properties of formation oil. At present, studies have investigated the influence of CO_2 on crude oil's physical properties [7–9] and realized that CO_2 has a low critical point and is easy to compress. It improves the formation oil property and thus improves oil recovery as it dissolves into crude oil to expand the volume of oil and reduce viscosity and interfacial tension [10,11]. The changes in physical property parameters such as crude oil volume coefficient, thermal expansion factor, and compression coefficient under different conditions are considered in these studies [12–14]. But these studies seldom consider the influence of methane mixing. Many scholars use numerical simulation software to explore the phase behavior changes of crude oil under different CO_2 concentrations and injection amounts [15], while these studies were not confirmed by experiments. It is also common to study the phase characteristics by combining experiments with equilibrium equations. Ruan Hongjiang [16], Zuo Mingsheng [17], Song Zhaojie [18] and others used the thermodynamic model of two-phase phase equilibrium to clarify the mass transfer law of crude oil–CO_2 under the influences of different factors. The experimental scheme is deficient. C. Ariza-Quiroga, et al. [19] paid attention to the changes in phase behavior and physical properties of crude oil under different conditions by combining experiments and prediction methods. In addition, it has great potential to study the microscopic interaction mechanism between CO_2 and crude oil by using molecular dynamics [20]. They all lack an understanding of the influence of mixed gas on the physical properties of crude oil. The introduction of mixed gas containing methane makes the change of physical properties of crude oil more complicated, which is closer to the actual application under the consideration of different concentrations and injection quantities. In addition, for the Bohai Oilfield, the effect of CO_2 + CH_4 mixed gas on oil's physical properties has hardly been reported.

In this paper, dehydrated crude oil and natural gas samples from the BZ 25-1 block were used to prepare simulated formation oil and high-pressure physical property test experiments were carried out to investigate the impacts of mixed gas on oil property. It provides a reference for the mechanism analysis and scheme optimization design of CO_2 flooding and storage in this block and similar reservoirs.

2. Experimental Description

2.1. Experimental Installation

The equipment used in the experiments mainly includes a sample container, PVT cell, viscometer, gas meter, plunger pump, balance, oven, piston container, vacuum pump, etc. The flow is shown in Figure 1. The parameters of the main apparatus are the following:

(1) PVT cell: TC-II-70 type high temperature and high-pressure formation fluid property analyzer with an operating pressure of 0–70 MPa, an operating temperature of room temperature −200 °C, and a maximum volume of 316 mL.

(2) Viscometer: HXND-2 falling ball viscometer with an operating pressure of 0–50 MPa and an operating temperature of room temperature −200 °C.

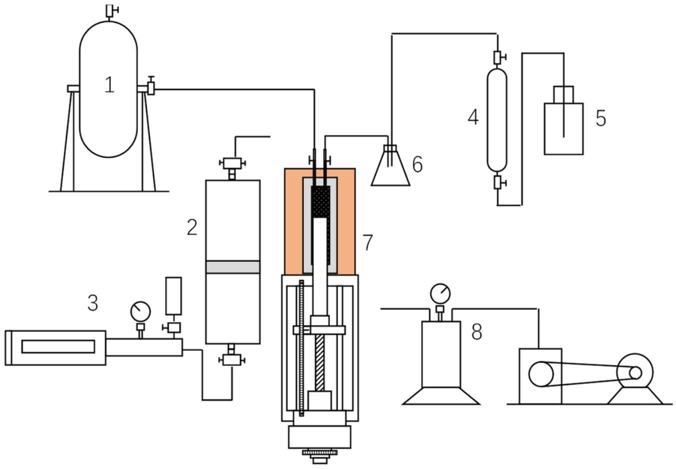

Figure 1. Equipment used to test parameters of formation oil: 1—viscosity meter; 2—sample container; 3—injection pump; 4—gas bottle; 5—balance bottle; 6—oil and gas separation bottle; 7—PVT cell; 8—vacuum system.

2.2. Experimental Samples

Oil from the SA3 section of the BZ25-1 reservoir was used, and the property parameters of the original formation oil are shown in Table 1. The molar mass of the degassed oil is 236.8, and its density is 0.8823 g·cm^{-3}. The solution gas used in the experiment was prepared according to the actual solution gas components, which are shown in Table 2. The dehydrated oil sample and prepared gas sample are mixed to prepare the simulated gas-containing formation oil, as shown in Table 3. CO_2 purity was adjusted by mixing with CH_4. The purity of CO_2 was defined as the mole percentage of CO_2 to the total of CO_2 and CH_4.

Table 1. Properties of BZ25-1 original formation oil.

Layer	**Reservoir Pressure /MPa**	**Oil Layer Temperature /°C**	**Saturation Pressure /MPa**	**Gas–Oil Ratio /m^3·m^{-3}**	**Volume Coefficient at Formation Pressure**	**Underground Crude Oil Density /g·cm^{-3}**	**Ground Crude Oil Density /g·cm^{-3}**	**Viscosity under Formation Pressure/mPa·s**
SA3	51.0	127.0	17.0	89.0	1.29	0.744	0.870	1.13

Table 2. Components of BZ25-1 natural gas.

Layer	**Relative Density**	**Components of Natural Gas/%**							
		CH_4	**C_2H_6**	**C_3H_8**	**C_4H_{10}**	**C_5H_{12}**	**C_6H_{14}**	**N_2**	**CO_2**
SA3	0.735	75.5	11.24	3.85	1.03	0.2	-	1.51	6.64

Table 3. Components of simulated formation oil.

Component	**Percentage/mol%**	**Component**	**Percentage/mol%**	**Component**	**Percentage/mol%**
CO_2	3.734186	nC12	1.123333	nC25	0.639754
N_2	0.566727	nC13	1.171444	nC26	0.485013
nC1	53.9555	nC14	1.385151	nC27	0.438839
nC2	10.53491	nC15	1.518425	nC28	0.395537
nC3	3.943798	nC16	1.360843	nC29	0.348941
nC4	3.229567	nC17	1.214582	nC30	0.280766
nC5	1.164507	nC18	1.024728	nC31	0.207071
nC6	0.621071	nC19	1.010208	nC32	0.199959
nC7	1.147974	nC20	0.947291	nC33	0.146798
nC8	1.157135	nC21	0.827465	nC34	0.122372
nC9	0.946416	nC22	0.775117	nC35	0.06786
nC10	0.98429	nC23	0.72877	nC36	0.026
nC11	0.951419	nC24	0.616234	nC37+	4.300913

2.3. Experimental Process

In order to investigate the impacts of mixed gas on the physical properties of formation oil, the experiment of different gas added ratios (percentage of mole of injected gas to mole of formation oil) was carried out. The saturation pressure, dissolved gas–oil ratio, viscosity, volume coefficient and expansion rate of the formation oil before and after injecting different amounts of gas were measured, and the change curve of each parameter with the gas injection amount was drawn to analyze the influence law.

The experimental process is shown in Figure 2, which mainly includes the following steps:

(1) Prepare the simulated formation oil sample with dehydrated crude oil and natural gas in the sample container;
(2) Transfer the oil sample from the sample container to the PVT cell to test the physical property parameters of the oil sample before adding CO_2 + CH_4 mixed gas;
(3) Inject the designed amount of mixed gas according to the scheme, and increase the pressure at the experimental temperature to fully dissolve the mixed gas and oil sample;
(4) Test the physical property parameters of crude oil after adding the mixed gas.

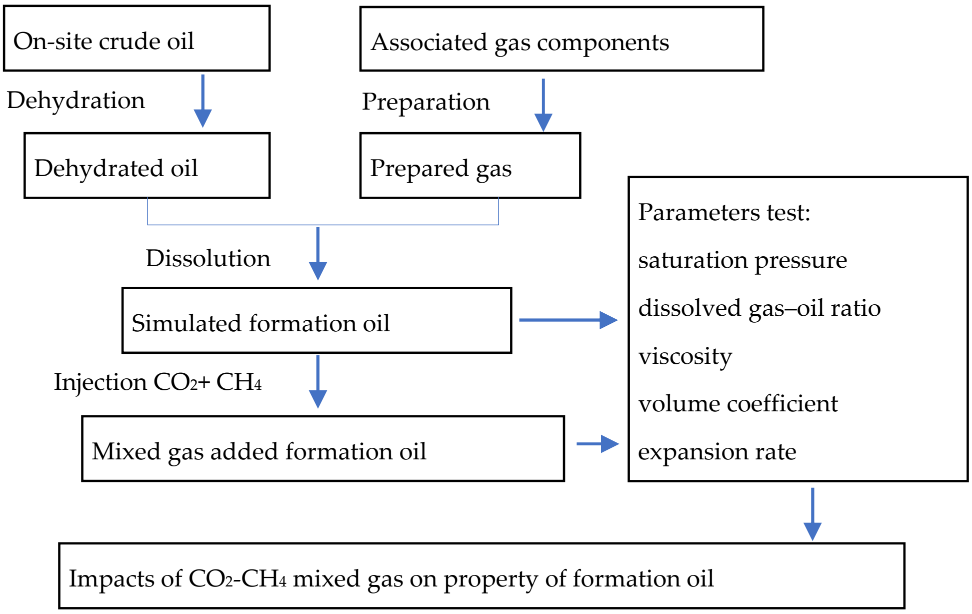

Figure 2. Experimental process.

The volume change of crude oil before and after gas injection and the volume change of crude oil under different pressures can be recorded by the PVT analyzer, and the expansion rate and saturation pressure can be obtained. The dissolved gas–oil ratio and volume coefficient can be obtained by single degassing with a gas meter, and, finally, the viscosity of the sample can be obtained by a viscometer, which is a commonly used experimental method for measuring high-pressure physical properties. All the experimental instruments used in the experiment are safe and reliable, and the experiment is carried out in strict accordance with the operating standards. The experimental process is safe and controllable, and the experimental data are true and fair.

3. Results and Discussions

At the temperature of 126.7 °C, the mixed gases with different CO_2 purity of 10%, 30%, 50%, 50%, 80% and 100% were injected, respectively, into the simulated formation oil, and the saturation pressure, expansion factor, dissolved gas-to-oil ratio (GOR) and volume coefficient were measured. The expansion factor here is defined as the ratio of the sample volume after gas addition to that before gas addition under the same pressure. The results are shown in Table 4. The changes in physical property parameters are shown in Figures 3–10.

Table 4. Properties of mixed gas-injected formation oil.

CO_2 Purity/%	Gas Added Ratio/mol%	GOR /$cm^3 \cdot cm^{-3}$	Saturation Pressure /MPa	Viscosity /mPa·s	Expansion Factor	Volume Coefficient
/	0	88.00	17.46	1.13	1.00	1.25
10	24.53	110.28	22.20	1.08	1.03	1.34
	46.83	130.12	26.50	1.03	1.06	1.39
	71.36	151.96	29.50	0.99	1.10	1.42
	80.28	160.32	31.00	0.98	1.12	1.43
30	30.11	115.20	21.90	1.05	1.04	1.38
	44.60	128.46	24.80	1.01	1.07	1.41
	72.64	154.25	28.70	0.94	1.11	1.45
	99.24	177.33	31.00	0.92	1.15	1.48
50	39.00	123.38	22.20	0.98	1.06	1.41
	62.44	144.23	25.50	0.89	1.11	1.45
	80.25	160.33	27.60	0.80	1.14	1.48
	105.92	182.96	29.60	0.75	1.17	1.50
80	34.55	119.32	21.80	0.98	1.06	1.41
	60.21	142.33	24.50	0.85	1.11	1.46
	86.97	166.27	27.30	0.75	1.16	1.50
	102.58	180.48	28.30	0.71	1.17	1.52
100	17.19	102.98	20.10	1.05	1.03	1.35
	35.1	118.65	21.50	0.95	1.07	1.41
	78.76	138.25	24.60	0.79	1.12	1.47
	97.78	175.23	27.60	0.68	1.18	1.53

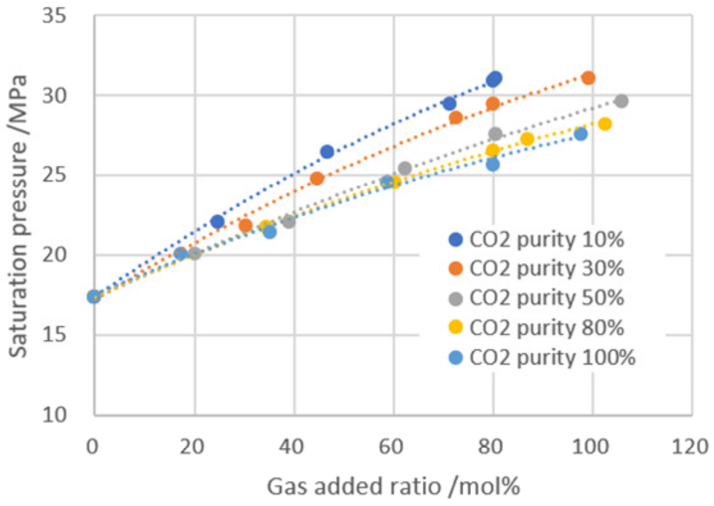

Figure 3. Changes of saturation pressure with gas injection amount.

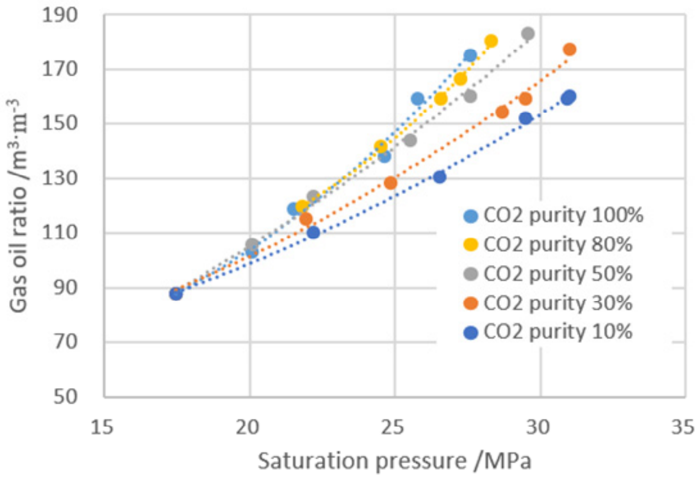

Figure 4. Changes of dissolved gas-oil ratio with saturation pressure.

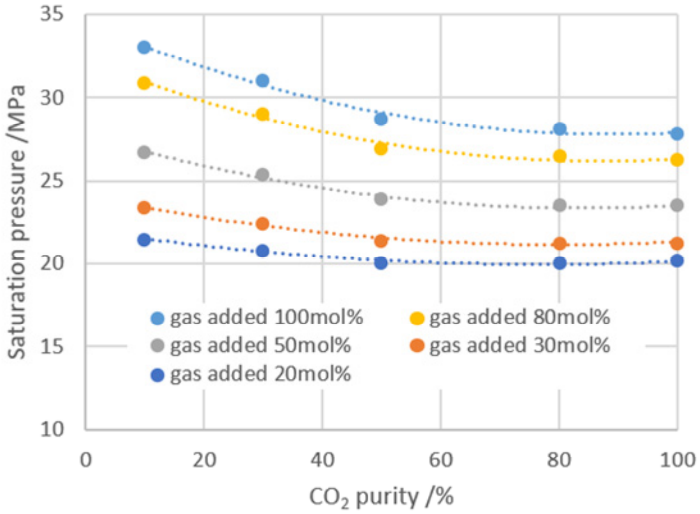

Figure 5. Changes of saturation pressure with CO_2 purity.

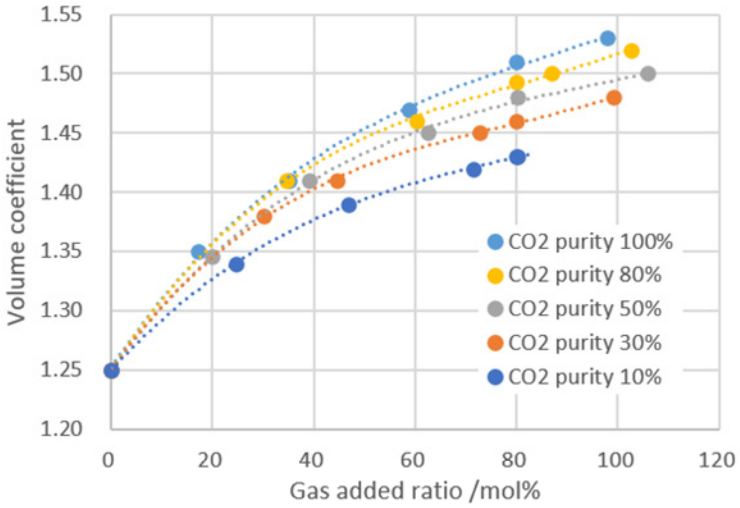

Figure 6. Change of volume coefficient with gas injection amount.

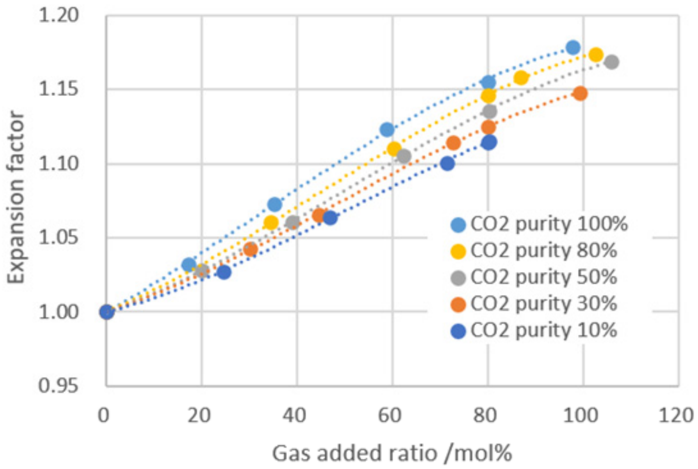

Figure 7. Changes of expansion factor with gas injection amount.

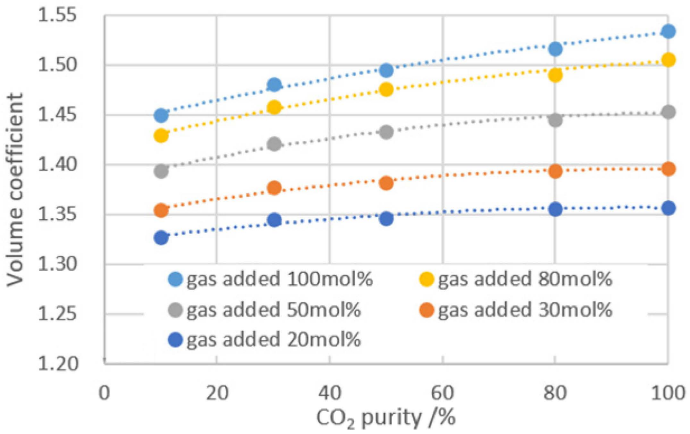

Figure 8. Changes of volume coefficient with CO_2 purity.

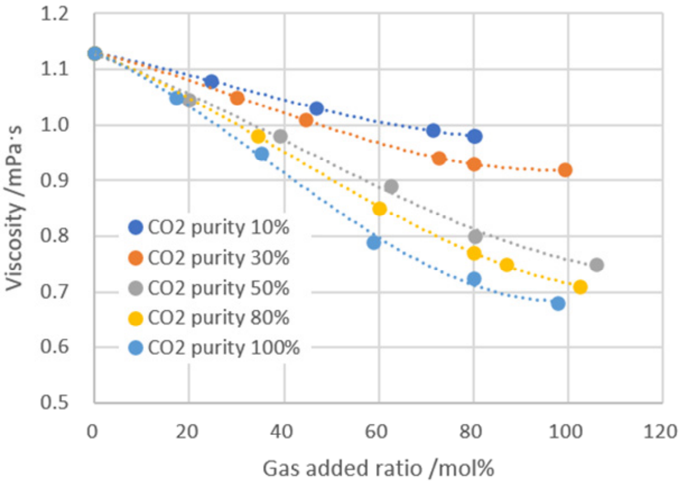

Figure 9. Changes of viscosity with gas added ratio.

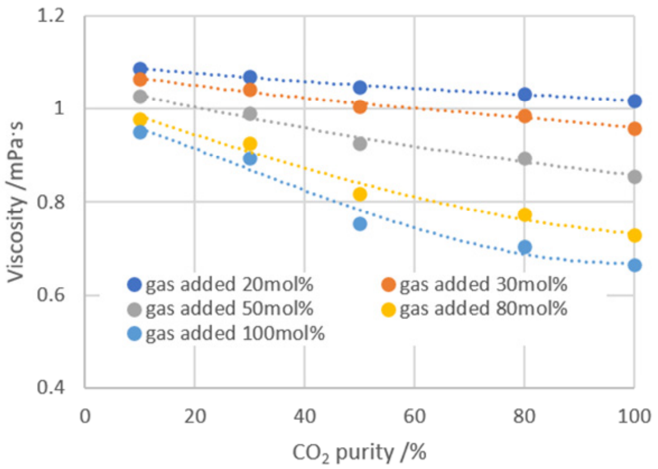

Figure 10. Changes of viscosity with CO_2 purity.

As can be seen from Figure 3, the saturation pressure of the system increases after gas injection. For the mixed gas with the same CO_2 purity, the more the gas injection amount is, the greater the saturation pressure of the system is. Increasing the gas injection volume can continuously increase the formation energy. When the gas injection amount is the same, the lower the purity of CO_2 is, the higher the CH_4 content in the mixed gas is, the higher the saturation pressure of the system, indicating that CO_2 is easier to dissolve into crude oil and CH_4 is more difficult to dissolve.

As shown in Figure 4, the higher the CO_2 purity, the more the gas dissolved in crude oil. For example, when the pressure is about 26 MPa, the dissolved gas–oil ratio of gas with CO_2 purity of 10% is about 130, while the dissolved gas–oil ratio of gas with CO_2 purity of 80% increases to about 160, the dissolved amount of gas increases by about 23%. Therefore, the greater the CO_2 content in the mixed gas is, the easier for the gas to dissolve into crude oil.

It can be more clearly seen from Figure 5 that when the injection amount is the same, the saturation pressure continues to decrease as the CO_2 purity increases, and the more the gas volume, the greater the decrease. For the gas added ratio of 100 mol%, CO_2 purity increased from 10% to 100%, and saturation pressure decreased from 33.0 MPa to 27.8 MPa, with a significant decrease in saturation pressure. The higher the purity of CO_2, the easier it is to dissolve and mix with crude oil. It can also be seen that when the CO_2 purity increases to about 60%, the saturation pressure of the system becomes gradually flat with the continued increase in the CO_2 purity.

The reason for this change is that CO_2 is easier to dissolve in crude oil than CH_4 under the same pressure. That is to say, for the same gas added ratio, the higher the CO_2 purity, the lower the pressure for the gas to completely dissolve. When the purity of CO_2 increases to more than 60%, the content of CH_4 in the mixed gas is relatively small, and the CH_4 content becomes less and less with the continued increase in CO_2 purity. Therefore, when the purity of CO_2 increases to more than 60%, the continued increase in CO_2 purity, the saturation pressure of the system gradually moves close to the saturation pressure of pure CO_2, and thus the saturation pressure of the system becomes flat gradually.

Therefore, for the reservoir injected with mixed gas to drive the oil, if the main purpose is to dissolve gas into the crude oil, the purity of CO_2 should be greater than 60%; if the aim is to increase the formation energy, the purity of CO_2 should be reduced, and the purity should be less than 60%.

It can be seen from Figures 6 and 7 that the volume coefficient and expansion factor of the formation oil increase with the increase in gas injection amount. For pure CO_2, when the injection volume increased from 0 to 100 mol%, the formation oil volume coefficient increased from 1.25 to 1.53, and the formation oil volume expanded by 18%. When the same gas volume is injected, the higher the CO_2 concentration, the larger the volume coefficient and the larger the expansion coefficient. The dissolution of the injected gas in the formation oil makes the volume of the formation oil expand, which helps the oil to discharge from the rock pores, which is beneficial to crude oil exploitation. Figures 6 and 7 also show that the increment of volume coefficient and expansion factor gradually slows down with an increase in gas injection. This is due to the pressure increasing with the injection volume, and the elevated pressure increases the compression effect on the system.

When the gas added ratio is small, the injected gas molecules enter the crude oil and fill the gap between the crude oil molecules. The gap between the crude oil molecules is not significantly increased, and the volume expansion of the system is small. With the increase in gas added ratio, the number of gas molecules dissolving into the crude oil molecules increases, the gap between the crude oil molecules is significantly increased, and the volume expansion becomes obvious. Therefore, with the relatively low gas injection ratio, the expansion factor curve is concave. While the gas added ratio increases to a certain value, the system volume continues to expand as the gas added ratio increases, but as a large amount of the injected gas is dissolved in the crude oil, the pressure must increase synchronously, and the increased pressure enhances the compression effect on the system. Therefore, at a higher gas added ratio, the expansion of the volume tends to be flat, and the expansion factor curve becomes convex.

CO_2 purity also has a clear effect on the volume coefficient and the expansion factor. Figure 8 shows that the volume coefficient and expansion factor increase significantly before CO_2 purity increases to about 60%, and after CO_2 purity exceeds 60%, the increments of volume coefficient and expansion factor decrease with CO_2 purity.

Figure 9 shows that the viscosity of the formation oil decreases with the increase in the gas injection amount. For pure CO_2, when the injection volume was increased from 0 to 100 mol%, the viscosity decreased from 1.13 mPa·s to 0.68 mPa·s, with a decrease of 39.8%. This shows that CO_2 dissolution in crude oil has a good viscosity reduction effect, which reduces the flow resistance of crude oil, improves the liquidity ability of crude oil, and is conducive to improving flooding efficiency. It can also be seen from Figure 8 that when the amount of gas injection exceeds 80 mol%, the increment of viscosity reduction of the formation oil continues to decrease with the amount of gas injection. From the perspective of viscosity reduction effect and gas injection cost, after the gas injection amount reaches 80 mol%, more gas injection is no longer suitable for the economic effect.

As can be seen from Figure 10, the influence of CO_2 purity on viscosity is quite obvious. With the same injection amount, the higher the CO_2 purity, the lower the system viscosity, indicating that CO_2 has a better viscosity reduction effect than CH_4. However, when the purity increases to 60%, the change of the viscosity reduction slows down as the purity continues to increase.

Considering the above experimental results, it can be known that the gas dissolved into crude oil can improve the physical properties of crude oil. With the gas addition, the volume of crude oil expands, and the viscosity of crude oil decreases. The more the gas is dissolved, the greater the physical improvement is. However, after the dissolution amount of gas increases to a certain extent, for more gas to be dissolved, the dissolution pressure must be increased, and the compression effect of the pressure increase on the system will be enhanced. From the effect of gas dissolution to improve the physical properties of crude oil, the gas injection amount is about 80 mol%. CO_2 purity has an obvious impact on the saturation pressure and the improvement degree of crude oil's physical properties. High-purity CO_2 is beneficial to improving the physical properties of crude oil, low-purity CO_2 can play the role of CH_4 in gas to improve the formation energy, and 60% CO_2 purity can better balance the improvement of crude oil's physical properties and improve the formation energy.

4. Conclusions

CO_2 flooding can improve the effect of crude oil exploitation and realize the geological CO_2 storage. CO_2 oil flooding and storage technology are important ways to achieve the goal of “carbon peak, carbon neutral”, which has good economic and social benefits. The high-pressure physical properties of CO_2-injected formation oil are the basis for analyzing the mechanisms of CO_2 displacement and storage and optimizing its technical scheme. For offshore fields, when CO_2 flooding is conducted, the associated gas produced by oil wells is mostly handled by local reinjection after mixing with CO_2. In order to carry out the CO_2 flooding and storage project in the BZ25-1 block in the Bohai oilfield, it is necessary to understand the effect of CO_2 addition and purity (mixed with CH_4) on the properties of formation oil. Experiments with simulated formation oil reveal the impacts of CO_2-CH_4 mixed gas on oil properties:

(1) With the increase of gas injection, the saturation pressure and dissolved gas–oil ratio, increases the volume coefficient and expansion rate, and reduces the viscosity, which is helpful in improving the exploitation of crude oil.

(2) Increasing the dissolution pressure can increase the dissolution amount of gas into formation oil; on the other hand, the pressure increase enhances the compression effect on the system. From the effect of gas dissolution to improve the physical properties of crude oil, the suitable gas injection amount is about 80 mol% for the BZ 25-1 block.

(3) High-purity CO_2 is beneficial to improving the physical properties of crude oil, while mixing with CH_4 can elevate the formation energy, and CO_2 purity of 60% can well balance the considerations on improving the oil properties and increasing the formation energy.

The injection of mixed gas has a great application scene in improving the physical properties of crude oil and replenishing formation energy. The cost can be effectively reduced and carbon sequestration can be effectively realized by reinjection of CO_2 and associated gas. Subsequent research can explore the long-term impact of mixed gas injection and the economic impact of different gas components.

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Data Availability Statement: The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

Conflicts of Interest: Author Renfeng Yang was employed by the company CNOOC Research Institute. 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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