Experimental investigation of large volume CO₂ injection for EOR and sequestration purposes

Qi Huan¹², Li Yiqiang^{12*}, Liu Zheyu¹²

1 State Key Laboratory of Petroleum Resources and Prospecting, China University of Petroleum (Beijing), Beijing 102249, China.

2 Petroleum Engineering Institute, China University of Petroleum (Beijing), Beijing 102249, China. (*Corresponding Author: Yiqiangli@cup.edu.cn)

ABSTRACT

The idea of implementing CO₂ injection for reservoir development is shifting from the traditional scheme of injecting small amounts of gas to prevent gas breakthrough, to a large pore volume (PV) injection approach based on expanding the swept volume gradually. To investigate the law and mechanism of EOR and gas sequestration by large PV injection during the CO₂ drainage method, a series of large PV CO₂ injection experiments were carried out. The cores' permeability and porosity at different stages of injection were measured, and then the microscopic characteristics and mineral types were analyzed, full-diameter cores were used for large PV CO₂ injection experiments to analyze the dynamic characteristics of CO₂ injection under the influence of factors such as the degree of miscibility, heterogeneity, and core dip angle. Results showed that with the progress of CO₂ injection, the porosity and permeability of the core followed a dynamic law of decreasing first and then increasing. The reaction of calcite, potassium feldspar, and other minerals with acid solutions to form precipitation and segregation, and the loss of fines leading to partial blockage of the throat, both are the main causes of early changes in the physical properties. However, migration and decomposition of the blockage and the development of micro-fractures caused by large PV CO₂ injection can further improve the physical properties. Furthermore, it is found that heterogeneity can easily cause premature CO₂ gas channeling and reduce the degree of recovery before the gas breakthrough. However, the high degree of miscibility is favorable for improving this situation. The amount of CO₂ storage per unit of crude oil produced shows a pattern of rapid decrease followed by a gradual increase throughout the whole drainage stage, and the gas injection volume remains essentially unchanged when it exceeds 2 PV. Therefore, achieving high recovery efficiency and CO_2 utilization rate is the fundamental reason for implementing large volume CO_2 injection. **Keywords:** CO_2 EOR-sequestration, large volume injection, miscible and immiscible, core flooding, physical property change, carbon neutrality.

1. INTRODUCTION

By making full use of the miscibility, gravity displacement, viscosity reduction, and oil swelling^[1-3], CO₂ can be injected into oil reservoir to EOR and storage of greenhouse gas at the same time^[4], realizing the winwin situation of oil production and promote carbon neutrality scenario^[5]. Since the 1950s, the United States had a lot of experience with enhanced oil recovery and storage of CO₂, where the recovery factor (RF) is enhanced by CO₂ flooding as tertiary recovery technology by 7–25 percentage points, with an average of 12 percentage points^[6-7]. In China, it was believed in the early potential evaluation stage that the overall increase of RF by miscible gas flooding such as CO2 flooding could reach 18.7 percentage points^[8]. A large number of field tests have shown that no matter whether it is CGI (continue gas injection) or WAG (water alternate gas), gas will float up and liquid will sink down due to gravity segregation, there will lead to a large unswept region. Once the gas is breakthrough, many fields will shut in wells because they do not have enough capacity to handle the gas produced and the corrosion challenges, this time the injection volume may be only 0.2-0.6 PV, and a lot of oil will remain in the reservoir^[9].

There comes a new idea that HCPV is positively related to the EOR according to several CO_2 flooding projects with high miscibility. The pilot test of CO_2 injection with small spacing in the North H79 block of the Jilin Oilfield showed that a total of 35×10^4 t CO_2 had been injected, the stable production period was about 4 years, the stable recovery rate was about 2.8%, and the RE was

[#] This is a paper for International CCUS Conference 2023 (ICCUSC2023), April 14-15, 2023, Beijing, China.

19.1%^[10]. If CO₂ was continuously injected to three times of hydrocarbon pore volume (3 HCPV), the RE of CO₂ flooding could reach 26.3%, which has great potential to EOR ^[11]. The results of the field test show that the oil production peak can be reached at the moment of gas breakthrough, and the recovery degree can continue to be enhanced due to diffusion and extraction. The idea of implementing CO₂ injection for reservoir development is shifting from the traditional scheme of injecting small amounts of gas to prevent gas breakthrough, to a new injection strategy: large pore volume (PV) injection approach based on EOR and CO₂ storage.

However, the dynamic change characteristics of displacement and storage efficiency, and the dynamic change law of core physical properties after large volume CO_2 injection have not been clarified at present. In this paper, the feasibility of large PV injection of CO_2 has been experimentally verified from micro and macro aspects, in order to enrich the theory of CO_2 flooding and geological sequestration under the background of carbon neutralization.

2. MATERIALS AND METHODS

2.1 Cores preparation

Two natural cores have been collected from Qingshui Subsag of the Liaohe oil field, the cores represent the typical properties of the actual reservoir. The cores were polished, oil washed, and dried at 85 °C for 48 h, the basic physical properties and mineral composition were shown in Table 1. Heterogeneous artificial full-diameter cores were designed according to the depositional and the physical property characteristics of the target reservoir, the diameter of each core was 10 cm and the length of the core was 30 cm, the average permeability of the core was 4 mD, the design and physical diagram were shown in Fig. 1.



Fig. 1. Heterogeneous artificial full-diameter core design diagram (a), and the physical diagram (b)

2.2 Nature cores after large volume CO₂ injection

The conventional core physical properties analysis method can not simultaneously reveal the effects of fluid

reaction with rock and long time gas flushing on rock physical properties. Therefore, this part of the experiment was designed to characterize the timevarying effects of two kinds of effects on core physical properties, the procedures as the following:

Table 1 Physical properties of natural core

No.	K (mD)	Ф (%)	L (cm)	Mineral content, %				
				Quartz	Calcite	Potassium feldspar	Plagioclase	Clay
1	2.7	15.23	4	49.3	1.2	9.3	33.5	6.7
2	1.8	15.14		55.9	0.5	3.6	33.2	7.3

1. Divide each core in half, continued to be evacuated for 6 hrs, then saturated with deionized water.

2. Set the experiment temperature and pressure as 110 $\,^\circ\!C\,$ and 35 MPa.

3. After the slow injection of 1 PV CO_2 , soaking for 7 ds, half of the core was removed for analysis (SEM and XRD).

4. 50 PV CO_2 was injected and the remaining half was removed after 7 ds soaking for next analysis.

2.3 Heterogeneous full-diameter cores flooding

1. As shown in Fig. 2, the full-diameter core was placed in core holder and continued to be evacuated for 48 hrs, then saturated with formation water and use dead oil to drive water to establish irreducible water.

2. Set the experiment temperature to 110 $^{\circ}$ C, the back pressure was set to 35 MPa and 25 MPa respectively to mimic miscible and immiscible flooding conditions.

3. Set the core holder angle to 10 $^\circ$ and the dynamic characteristics of oil and gas production were recorded and analyzed.

3. RESULTS AND DISCUSSION

By following the above procedures, the physical properties of the cores and the CO₂ displacement behavior in heterogeneous cores were analyzed and studied. The results are discussed in the following steps.

3.1 Physical properties change after large volume CO₂ injection

Large volume injection of CO_2 into reservoirs can result in changes in physical properties, which has been proven in many field practices. CO_2 has a high solubility in formation water, and the CO_2 formed in water will ionize a large number of H⁺, $CO_3^{2^-}$, and HCO_3^{-} , which will form precipitates with Ca^{2+} and Mg^{2+} in the original formation water. It will also reduce the PH of formation





water and react with minerals to dissolve and mineralize. generating new substances. This is also one of the mechanisms of CO₂ permanent storage in underground porous media^[12]. In order to explore the changes of reservoir physical properties under the coexistence of CO₂ scour and mineralization reaction, deionized water was selected as the experimental water in this experiment, so as to exclude the influence of ions in formation water on the experimental results. Fig. 3 shown the physical properties change after 1PV volume CO₂ injection, it can be known that the porosity and permeability both decreased after 1 PV CO₂ flooding. The No.1 core had a 14% decrease in permeability and a 0.5% decrease in porosity, while the No.2 core had a 21.2% decrease in permeability and a 3.6% decrease in porosity. It was found that the pore was blocked by new minerals and clay particles (Fig. 5. (a)), which was the fundamental cause of the core porosity and permeability decrease from the Fig. 3. Moreover, XRD results showed that the content of calcite, potassium feldspar, and chlorite decreased, while the content of kaolinite increased significantly. After 50 PV CO₂ injection, in the process of displacement, both porosity and permeability decrease first and then increase. The permeability increased and was higher than the initial state, porosity increased slightly but less than the initial state. In general, after 50 PV CO₂ injection, the permeability of the No.1 core increased by 11.11%, and the porosity decreased by 12.33%, while the permeability of the No. 2 core increased by 3.03% and the porosity decreased by 9.4%. as shown in Fig. 4. As shown in Fig. 5 (b), after large PV CO₂ injection, partially blocked pores were unblocked, and the developed small fractures continue to extend and expand along the initial strike, enhancing the conductivity of the fractures. The content of feldspar and clay further decreased, CO₂ erosion resulted in

microfracture expansion, and surface dissolution of mineral particles. The fundamental reasons for the change of physical properties are the dissolution, precipitation, and micro-fracture development caused by the mineralization reaction during large volume CO_2 injection. Therefore, changes in porosity and permeability caused by mineralization should be taken into account in CO_2 flooding and reservoir engineering design.



Fig. 3. Changes in porosity and permeability of natural core after 1 PV \mbox{CO}_2 injection

3.2 Dynamic characteristics of CO₂ flooding

The results show the recovery factor and the production rate change during the CO_2 flooding as shown in Fig. 6. For the miscible displacement process, the final recovery factor after 3 PV displacement was 61.69%, much higher than the immiscible displacement recovery factor of 34.87% after 3 PV displacement. Moreover, the gas time of miscible displacement was 0.37 PV, and the

recovery degree before the gas breakthrough was 28.3%, which could still greatly enhance the recovery degree after the gas breakthrough.





The stage after the gas breakthrough was the important oil recovery stage of miscible flooding, while the gas breakthrough time of immiscible flooding was 0.24 PV, the recovery factor before the gas breakthrough was 13.8%, and the recovery factor after the gas breakthrough was 21.07%. After 1.5 PV displacement, immiscible CO_2 flooding was difficult to displace oil, but oil was still displaced after 2 PV miscible displacement. Therefore, the oil recovery of miscible flooding was always higher than that of immiscible flooding as shown in Fig. 7. The gas breakthrough time was the highest oil production rate. Compared with immiscible flooding, miscible flooding results in late gas breakthrough and a higher ultimate recovery factor.

Moreover, it shows that before gas breakthrough, the production oil's color was black, same as the original



Fig. 5. Microscopic characteristics after 1 PV CO₂ injection (a), and microscopic characteristics after 50 PV CO₂ injection (b)

, when gas was breakthrough, the liquid production rate was high, and the color of oil was lighter than before, at the end of the drainage process, there was little oil film that could be produced, almost the lighter component, which improved the volume displacement and oil expansion were the main EOR mechanisms before gas breakthrough, while diffusion and extraction were the main EOR mechanisms after gas breakthrough.





3.3 CO₂ storage characteristics

At present, there are few theoretical calculation methods for CO_2 storage, and it is difficult to apply reservoir engineering methods on a laboratory scale. Therefore, a new theoretical calculation method and experimental method of CO_2 storage are proposed, CO_2

storage efficiency was calculated by accurately measuring the amount of CO₂ injected and produced, the calculation method is shown in Eq. 1. The CO₂ storage efficiency is defined as the ratio of the amount of CO₂ stored in the pore to the total amount of CO₂ injected, CO₂ utilization efficiency is defined as the amount of CO₂ stored to produce a unit mass of crude oil. As shown in Fig. 8, before the gas breakthrough, the CO₂ storage efficiency of both flooding methods was 100%, with the increase of PV injected, the CO₂ storage efficiency showed a decreasing trend. Before 1 PV displacement, the CO₂ storage efficiency achieved by the two oil displacement methods was almost similar. After 3 PV displacement, the CO₂ storage efficiency achieved by miscible flooding was 39.46%, and that by immiscible flooding was 31.38%, which had a big difference.





The variation of CO_2 utilization efficiency with the injected PV was shown in Fig. 9. The CO_2 utilization efficiency showed a rapid decrease and a slow increase and then maintained a stable and constant dynamic change. After 3 PV displacement, CO_2 storage per unit crude oil production was 0.82 g/g, and that in unit crude oil production by immiscible flooding was 0.68 g/g. It can be considered that the implementation of CO_2 large PV injection can not only ensure a higher recovery factor but can also achieve higher CO_2 utilization. Moreover, this value can predict the CO_2 dynamic storage based on crude oil production in the actual production process.





4. CONCLUSIONS

With the progress of CO_2 flooding, the porosity and permeability of the core appear a dynamic change law of decreasing first and then increasing. The main reasons for the deterioration of physical properties are the blockage of pores caused by the dissolution of calcite, potassium feldspar, and chlorite minerals and clay particle loss, while the fundamental reasons for the improvement of reservoir physical properties are the migration of plugging materials and the development of micro-fractures caused by mineralization reaction during large volume CO_2 injection.

Reservoir heterogeneity is easy to cause premature gas breakthrough and reduces the degree of recovery before the gas breakthrough. Miscibility is conducive to reducing the adverse effects caused by heterogeneity and achieving higher recovery factor and CO_2 utilization is the fundamental reason for the implementation of large PV CO_2 flooding.

Miscible displacement is the basis for gas flooding to greatly enhance oil recovery, the technical approaches to realize the limit recovery factor are to design a large PV injection scheme, strengthen miscibility and expand swept volume continuously.

ACKNOWLEDGEMENT

We would like to thank the China Liaohe Oilfield of CNPC and the China University of Petroleum (Beijing) for their support of this work, the anonymous reviewers are also sincerely thanked.

DECLARATION OF INTEREST STATEMENT

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. All authors read and approved the final manuscript.

REFERENCE

[1] ROGERSON, A.; MEIBURG, E. Numerical simulation of miscible displacement processes in porous media flows under gravity. Physics of Fluids A: Fluid Dynamics, 1993, 5.11: 2644-2660. (Reference to a journal publication)

[2] STALKUP, Fred I. Displacement behavior of the condensing/vaporizing gas drive process. In: SPE annual technical conference and exhibition. OnePetro, 1987.

[3] MOORTGAT, Joachim, et al. CO_2 injection in vertical and horizontal cores: Measurements and numerical simulation. SPE Journal, 2013, 18.02: 331-344. (Reference to a conference) [4] STALKUP, F. I. Carbon dioxide miscible flooding: Past, present, and outlook for the future. Journal of Petroleum Technology, 1978, 30.08: 1102-1112. (Reference to a journal publication)

[5]SAMBO, Chico, et al. A Technical Review of CO₂ for Enhanced Oil Recovery in Unconventional Oil Reservoirs. Geoenergy Science and Engineering, 2023, 221: 111185. (Reference to a journal publication)

[6]WANG Gaofeng, QIN Jishun, SUN Weishan. CCUS cases analysis and industrial development suggestions. Beijing: Chemical Industry Press, 2020. (Reference to a journal publication)

[7] LI Shilun, SUN Lei, CHEN Zuhua, et al. Further discussion on reservoir engineering concept and development mode of CO_2 flooding-EOR technology. Reservoir Evaluation and Development, 2020, 10(3): 1–14. (Reference to a journal publication)

[8] SHEN Pingping, YUAN Shiyi, HAN Dong, et al. Strategy study and potentiality evaluation of EOR for onshore oil fields in China. Acta Petrolei Sinica, 2001, 22(1): 45–48. (Reference to a journal publication)

[9] STALKUP F I, STEIN M H, LAKE L W, et al. CO₂ flooding. Texas: Society of Petroleum Engineering, 1998. (Reference to a book)

[10] LIAO Guangzhi, HE Dongbo, WANG Gaofeng, et al. Discussion on the limit recovery factor of carbon dioxide flooding in a permanent sequestration scenario. Petroleum Exploration and Development, 2022, 49(6): 1262-1268. (Reference to a journal publication)

[11] HU Yongle, HAO Mingqiang, CHEN Guoli, et al. Technologies and practice of CO_2 flooding and sequestration in China[J]. Petroleum Exploration and Development, 2019, 46(4): 716-727. (Reference to a journal publication)

[12] MATTER, Jürg M.; KELEMEN, Peter B. Permanent storage of carbon dioxide in geological reservoirs by mineral carbonation. Nature Geoscience, 2009, 2.12: 837-841. (Reference to a journal publication)