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Experimental Verification of Manmade Gas Hydrate Reservoir System for Sub-seabed CO_2 Sequestration#

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ABSTRACT

Sub-seabed sequestration of CO_2 is a potential method for reducing the atmospheric level of greenhouse gas. The geological exploitation in sub-seabed hydrate zone shows that free gas could be sealed beneath it, which provides a feasible idea for CO_2 sub-seabed sequestration. In this study, the CO_2 was injected into the water-containing porous media to simulate the CO_2 sub-seabed sequestration process, and the magnetic resonance imaging (MRI) technique was employed to recognize the real-time reservoir state. The CO_2 hydrate formed during the CO_2 upward migration process and produced sealing effect for external fluids. In addition, it was found that at least 8 MPa of overpressure can exist stable beneath the CO_2 hydrate cap, which ensures the security of CO_2 marine sequestration and increases the CO_2 sequestration depth. These results indicate that the hydrate cap provides a feasible and safe method for CO_2 geological sequestration, and it is significant to promote the potential of sub-seabed CO_2 sequestration.

Keywords: CO_2 sequestration, hydrate cap, sealing effect, MRI

NONMENCLATURE

Abbreviations	
MRI	Magnetic Resonance Imaging
CCS	CO_2 Capture and Storage
MI	Mean Intension
Symbols	
S_{wi}	1

1. INTRODUCTION

CO_2 capture and storage (CCS), which is regarded as the leading option to mitigate global warming, refers to the technologies that capture CO_2 at some stage and store it [1, 2]. CCS includes CO_2 sequestration, and CO_2 sequestration refers to the direct injection of CO_2 into specific geological structures, which can be divided into CO_2 geological sequestration and CO_2 marine sequestration [3]. CO_2 geological sequestration requires a stable environment condition, a cap layer, and sufficient storage capacity, which mainly includes saltwater, oil and gas fields, mineral sequestration, and coal sequestration. There are certain disadvantages to geological sequestration: Saltwater layer has strict requirements on geological structure; The main purpose of oil and gas field sealing is to make profits; Gas layer sequestration will waste coal resources, and mineral sequestration will cause environmental problems [4, 5]. As for CO_2 marine sequestration, it refers to injecting the captured CO_2 into the ocean, where the CO_2 is dissolved in seawater or exists as a liquid carbon lake [5]. However, Injecting carbon dioxide directly into the ocean may change the marine environment and harm marine life.

In specific seabed environments, CO_2 hydrate forms when CO_2 gas comes into contact with water, and CO_2 sequestration through hydrate formation in seabed sediments is also a promising and feasible method [6]. The stored CO_2 volume in hydrate phase is more than 100 times that compared with a unit volume of hydrate [7]. More recently, research found that hydrate formation in sub-seabed sediments can lead to rapid permeability reduction, and free gas can be trapped below it [8]. In this study, CO_2 gas was injected into the water-saturated reservoir to simulate CO_2 sequestration in sub-seabed, the state of the reservoir was real-time observed by MRI technology, and the influence of different reservoir pressures on CO_2 sequestration was explored. The results provide the important basis and location selection for CO_2 sub-seabed sequestration.

2. MENTALITY OF SUB-SEABED CO_2 SEQUESTRATION

1 volume of pore space in marine sediment can sequestrate 160 volumes of CO_2 (in standard conditions) by hydrate formation [9], the sequestration capacity of injecting CO_2 directly into sub-seabed sediments is larger. However, the method of injecting CO_2 directly requires a cap to prevent leakage. The Sleipner storage project, which is the first commercial-scale CO_2 injection project in the world, the CO_2 was injected into a saline aquifer found between 800 and 1000 m below the seafloor, and the CO2 is prevented from escaping to the surface by a caprock layer [10]. However, this caprock layer is to be met rather than sought. Fortunately, hydrate formation in pore space of sediments results in a great decrease in reservoir permeability and blockage of CO_2 leakage [11]. This zone where hydrate formation is regarded as the hydrate cap, then, the CO_2 could be injected below it to sequestrate, as shown in Fig. 1. Injecting CO_2 into the sub-seabed sediments, where the temperature and pressure are in the thermodynamically stable region of CO_2 hydrate. Hydrate forms during the CO_2 upward escape process and forms the hydrate cap with plugging effect. The CO_2 is further injected below this hydrate cap for sequestration. The small amount of CO_2 leaked before the full formation of hydrate cap will dissolve in the overlying seawater.

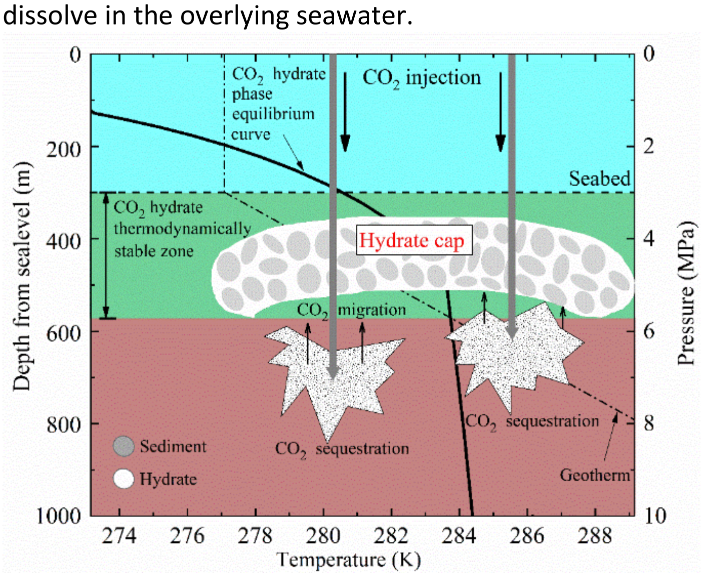

Fig. 1 Sub-seabed CO2 sequestration beneath the hydrate cap

3. CAPILLARY SEALING EFFECT OF CO_2 HYDRATE ZONE

Suitable geological conditions for CO_2 hydrate formation above the CO_2 injection area are vital conditions for sub-seabed CO_2 sequestration [12]. In this study, the CO_2 sequestration under two different pressure conditions was investigated. The temperature of reactor was kept constant at 274.15 K, and the back pressure was consistent at 2.0 MPa for Case 1, and 2.5 MPa for Case 2, which were all in the thermodynamical area of CO_2 hydrate. The CO_2 was injected from the bottom of reactor at the flow rate of 4ml/min to simulate the CO_2 upward leakage process after CO_2 was injected into sub-seabed. After the formation of hydrate cap, the N_2 was used to pressurize and verify the stability of hydrate cap.

3.1 Formation of CO_2 hydrate cap

Fig. 2(a) demonstrates the CO_2 leakage process after injecting CO_2 into sub-seabed. The BZ-01 glass beads with a porosity of 34.8% were used as sediments, the reactor was in the state of saturation before CO_2 injection. The pressure above the reactor (back pressure) was kept constant by high–precision syringe pump, the CO_2 was injected from the reactor bottom. In Case 1, the bottom pressure was consistent with back pressure before 48.5 min, and the moles of CO_2 leakage increased, as shown in Fig. 2(b). However, the CO_2 leakage stopped after 48.5 min and the bottom pressure increased, indicating that hydrate cap formed at 48.5 min. The CO_2 was continuously injected until the bottom pressure reached 6.0 MPa. The CO_2 liquefaction pressure was 3.57 MPa at 274.15 K, thus, the CO_2 was liquefied during this period. The decrease of the slop of bottom pressure curve was caused by CO_2 liquefaction. As the degree of liquefaction increased, the rate of bottom pressure increased until complete liquefaction, and caused the rapid increase, for the reason that gas has higher compressibility than liquids [13]. In Case 1, the hydrate cap with sealing effect for both gases and liquid CO_2 formed during the CO_2 upward leakage process, which provides the feasibility and security for sub-seabed CO_2 sequestration.

The N_2 was injected at the rate of 4 ml/min after the bottom pressure reached 6.0 MPa to verify the stability of CO_2 hydrate cap. Finally, the bottom pressure increased to 10.0 MPa (maximum pressure of reactor) and there was an 8.0 MPa pressure difference existed between the two ends of reservoir, indicating the hydrate cap can withstand overpressure of 8.0 MPa or even higher. Moreover, no gas leaked from the reactor after the formation of hydrate cap. Thus, injecting CO_2 beneath the CO_2 hydrate cap is a feasible method for CO_2 sequestration.

In Case 2, the back pressure was 2.5 MPa, the CO_2 stopped leaking from the reservoir after 12.1 min and the bottom pressure increased, indicating the hydrate cap formed at this time. Similar to Case 1, the CO_2 liquefied after the bottom pressure increased to 3.57 MPa. The

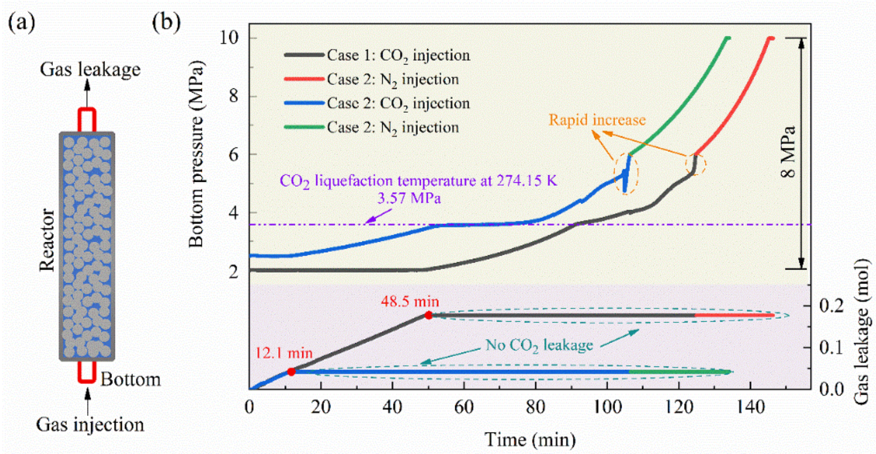

Fig. 2 Reservoir characteristics during CO2/N2

injection process: (a) Diagram; (b) Changes of bottom pressure and gas leakage

pressure drop before the rapid increase was caused by CO_2 liquefaction. The bottom pressure also reached 10.0 MPa after the N_2 injection. Different from Case 1, the time of hydrate cap formation was early than Case 1 and caused less leakage of CO_2 leakage, as shown in Table 1. Higher back pressure accelerated the CO_2 hydrate formation by enhancing mass transfer [14], which promoted the formation of hydrate cap and reduced the CO_2 leakage. Thus, the geological area with more favorable hydrate formation conditions was more suitable for CO_2 sequestration site.

Table 1 Summary of CO_2 hydrate cap in Cases 1 and 2

Case	Back pressure (MPa)	Formation time (min)	CO2 leakage (mol)	overpressure (MPa)
1	2	48.5	0.177	8
2	2.5	12.1	0.042	7.5

3.2 Characteristics of hydrate cap

Taking the advantage of pure phase encoding to identify the fluid phase distribution [15], the water distribution and phase transition in the reservoir were visually analyzed by magnetic resonance imaging system (MRI) system. As shown in Fig. 3, the MRI images darkened gradually indicating the decrease of water in FOV. The changes in water distribution could be divided into 4 stages: 0-4 min was the first stage, the water was displaced by CO_2 gas and caused the MRI image to darken at the middle of reactor; In stage 2, the bottom pressure was below the CO_2 liquefaction pressure, and the water consumption caused by hydrate formation darkened the MRI images; In stage 3, the CO_2 liquefied and caused water migration; In stage 4, the pore water was in a steady state although the bottom pressure increased continuously, indicating the hydrate cap was able to withstand high load pressure. Fig. 4(a) demonstrates the changes of water saturation in FOV, the water saturation at time i (S_{wi}) is calculated by $S_{wi} = I_i/I_{full}$, where I_{full} and I_i were MI signal of complete water saturation and water saturation at time i, respectively. The hydrate saturation is calculated by $S_h = 1.25 \times (I_0 - I_i)/I_0 \times S_{w0} \times 100\%$, S_{w0} is the water saturation at 0 min [16]. The water saturation decreased with the CO_2 and N_2 injection, which was consistent with the MRI images. The hydrate cap was constituted with the low hydrate saturation and high-water saturation, as shown in Fig. 4(b). The capillary sealing effect of hydrate cap was produced for the reason that local humps of hydrate saturation in high water saturation areas [17].

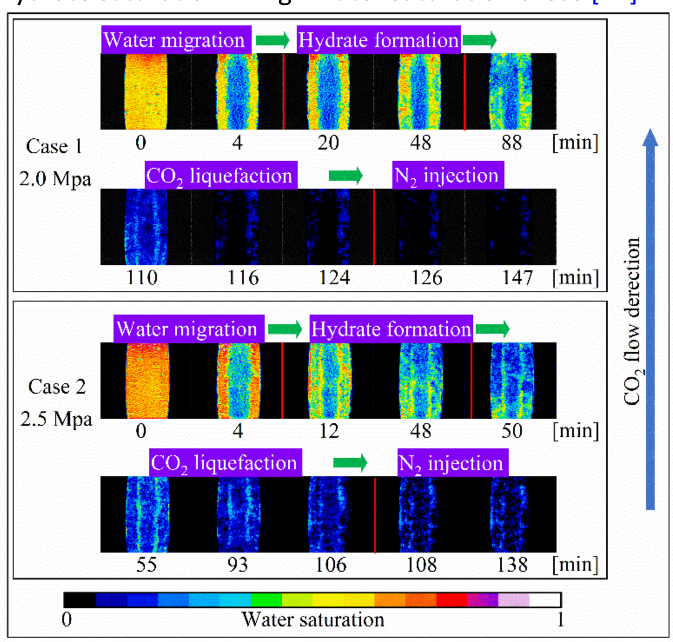

Fig. 3 Changes of water distributions during the injection process

3.3 Promoting of hydrate sealing zone on CO_2 sequestration

As mentioned above, there was at least 8 MPa overpressure can exist stable beneath the hydrate cap,

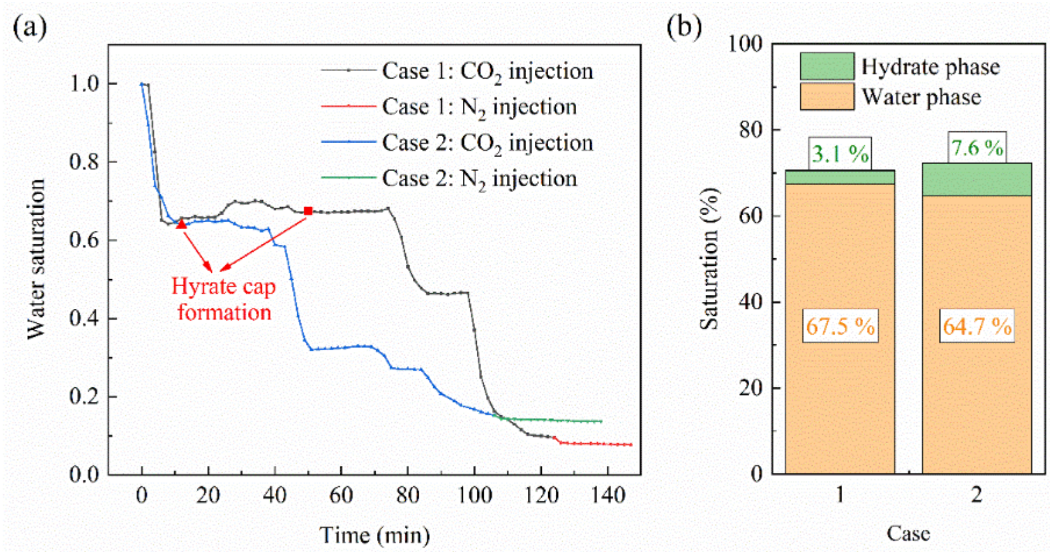

Fig. 4 Characteristics during the injection process (a) Water saturation in FOV; (b) Hydrate saturation and water saturation at the time of hydrate cap formation

which was independent of the pressure change caused by gravity. The high overpressure not only represented the safety of hydrate cap but also showed the great potential for CO_2 sequestration. The overpressure of 8.0 MPa is sufficient to prevent the sequestrated CO_2 break this capillary barrier, and this overpressure means the increase of CO_2 vertical storage space [18].

4. CONCLUSION

Injecting CO_2 into sub-seabed is a promising method for CO_2 sequestration, the upward leakage CO_2 gas induces hydrate formation in the thermodynamically stable conditions of CO_2 hydrate, and further forms the hydrate cap. Local humps of hydrate saturation in high water saturation area products capillary sealing effect for both gaseous and liquid CO_2. In addition, the hydrate cap can withstand the overpressure of 8.0 MPa or higher. The hydrate cap has high security for CO_2 sequestration. Moreover, the 8 MPa overpressure means the increase of CO_2 vertical storage space. Sub-seabed CO_2 sequestration beneath the hydrate cap is a safe, promising, and feasible solution to reducing the atmospheric level of the greenhouse.

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