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Development status and prospect of natural gas hydrate recovery by CO_2 replacement

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ABSTRACT

Using CO_2 replacement to extract natural gas hydrates can not only achieve CO_2 storage, but also ensure the safety of natural gas hydrate extraction, which is one of the necessary ways to achieve China's "dual carbon goals". However, the existing research on CO_2 replacement extraction of natural gas hydrates is not yet mature, making it difficult to support large-scale CO_2 storage and commercial extraction of natural gas hydrates. Therefore, this article systematically elaborates on the current situation and necessity of CO_2 storage, the distribution range and development challenges of natural gas hydrates in China, and analyzes the feasibility and wellbore integrity issues of CO_2 replacement extraction of natural gas hydrates. Research has shown that: CO_2 emissions are increasing year by year, seriously affecting the living environment of humans and other species, and must be sealed. China is rich in natural gas hydrates, but conventional mining can easily affect the stability of the strata and trigger natural disasters such as submarine landslides. Since single displacement reaction is a spontaneous process and CO_2 hydrate sediment intensity is higher, the feasibility of CO_2 replacement for natural gas hydrate exploitation is high. It is necessary to focus on the corrosion integrity of the cement and underground tubing during the replacement mining process.

Keywords: CCUS, natural gas hydrate, replacement mining, feasibility, corrosion integrity

1. INTRODUCTION

In nature, natural gas hydrates are mainly CH_4 hydrates, which are cage like crystalline compounds formed by the guest molecule CH_4 and multiple host H_2O molecules under certain temperature and pressure conditions[1]. 1m^3 of natural gas hydrate can decompose under standard conditions to produce 0.87m^3 of free water and 164.6m^3 of CH_4 gas, making it a rich source of natural gas[2]. As a clean energy source, natural gas hydrates are widely distributed in deep-water regions and polar permafrost regions of China. Promoting their commercial development is one of the keys to reducing China's energy dependence on foreign countries, achieving China's "dual carbon goals", and establishing China's international status as an energy powerhouse. The existing mining methods and their advantages and disadvantages are shown in Table 1[3],[4].

Tab. 1. Natural gas hydrate extraction methods

Method	Principle	Pros	Cons
Depressurization	Reduce the pressure of hydrate reservoir to the phase equilibrium pressure of hydrate	No energy loss, simple, easy to operate, and low cost	Slow production speed, easy to cause geological risks, ice blockage in the later stage of mining, hydrate regeneration, decreased gas production efficiency, and impact on reservoir stability

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Thermal activation	Injecting hot water, steam, or microwave into natural gas hydrate reservoirs	The operation of hot fluid injection is simple and the hot fluid can be recycled; High conversion rate of microwave thermal energy; Clean and environmentally friendly solar heating	The utilization rate of heat injection fluid capacity is low, suitable for hydrate extraction with high saturation; There are bottlenecks in microwave and solar heating technologies
Inhibitor injection method	By injecting chemical inhibitor into the gas hydrate bearing reservoir, its phase equilibrium curve is moved to high pressure and low temperature, promoting the decomposition of gas hydrate	Improve the driving force of hydrate decomposition without energy loss	High cost of inhibitors, low injection rate, poor economic benefits, environmental pollution, and impact on reservoir stability
Solid-state fluidization	Crush the hydrate containing reservoir into solid particles, then mix with seawater to form a slurry and transport it to the hydrate decomposition platform to extract natural gas, while returning the solid sediment back to the reservoir	By in-situ solid-state development, engineering geological hazards and greenhouse effect have been reduced, achieving controllable and orderly decomposition of hydrates within the closed wellbore range	Risk of erosion and wear of pipe columns, failure to meet commercial mining requirements
CO_2 replacement method	Injecting CO_2 gas into hydrate containing reservoirs can replace CH_4, and the heat released during the replacement process can promote the decomposition of natural gas hydrates	Ensuring reservoir stability, low water production, low energy consumption, achieving CO_2 storage, mitigating greenhouse effect, and promoting CCUS	Difficulty in CO_2 injection, limited displacement rate, low efficiency, cost, and low CH_4 recovery rate

It can be seen that CO_2 replaces CH_4 in natural gas hydrates to form CO_2 hydrates, which can not only extract CH_4 but also achieve geological storage of CO_2, in order to reduce the CO_2 content in the atmosphere. This is in line with China's goal of building a green country and is a highly promising mining method. Therefore, this article systematically elaborates on the current situation and necessity of CO_2 storage, as well as the difficulties in the development of natural gas hydrates in China. It analyzes the feasibility of CO_2 replacement for extracting natural gas hydrates, and finally proposes the wellbore integrity issues faced in the process of CO_2 replacement for CH_4. Research can provide reference for CO_2 replacement CH_4 technology.

2. CURRENT STATUS OF CO_2 STORAGE

By analyzing 4213 highly cited articles on carbon sequestration in WOS from 1950 to 2022, the changes in citation frequency and quantity over time are shown in Figure 1. The research on carbon dioxide storage technology started late and only received scholars' attention in 2011. Relevant papers began to increase, and 555 high-level articles were published in 2019, further proving the development prospects of carbon storage technology.

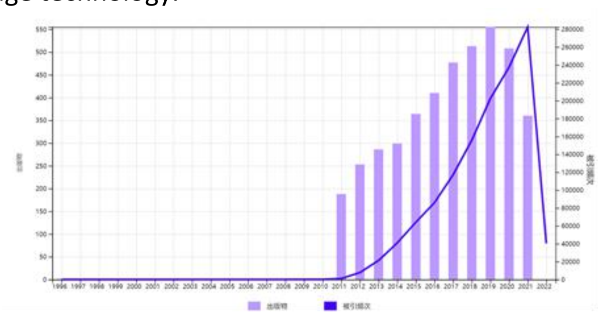

Fig. 1. Number and citation frequency of carbon sequestration related papers (data source: WOS)

According to data from the Global CCS Institute, as of the end of 2020, there were a total of 65 commercial CCUS projects worldwide that capture and permanently store approximately 40 million tons of carbon dioxide annually. Among them, 26 were operational, 2 were decommissioned, 3 were under construction, 13 were in the early stages of engineering design, and 21 were in the early stages of development[5]. Tab. 2 shows some CCS/CCUS projects, indicating that CCUS is a major trend in international energy development[6].

Tab. 2. Certain CCS/CCUS project

Project	Time	Idustry	Site	Amount of CO_2 captured/stored

Sleipner	1996	Statoil	Salt water layer Utsira sandstone in Sleipner gas field, Norway	100000 tons/a
InSalah	2004	Joint venture between BP, Sonatrach and Statoil	Salah gas field in central Algeria	120000 tons/a
Otway	2005	CO2CRC	Otway Basin in Victoria, Australia	—
Snahvit CCUS	2007	Equinor	Melkøya	700000 tons/a
China Shenhua Group CCS (Carbon Capture and Storage)	2010	China Shenhua Group	Yijinluo Banner, Ordos City, Inner Mongolia Autonomous Region	100000 tons/a
Boundary Dam	2014	SaskPower and the Government of Canada	Estwan, Saskatchewan, Canada	1000000 tons/a
Quest	2015	Shell, the Government of Canada, and the Government of Alberta	Saskatchewan, Alberta, Canada	1000000 tons/a
Petra Nova	2016	NRG Energy Company and JX Japan Oil and Gas Exploration	Jackson County, Texas	1600000 tons/a
Gorgon	2019	Chevron, Shell, ExxonMobil	Barrow Island, Australia	4000000 tons/a
CCSM Project, Saudi Arabia	2020	Saudi Aramco State owned oil company and Total Air Products	Eastern region of Saudi Arabia	800000 tons/a
Enping 15-1 Oilfield Group Offshore CCS	2021	China National Offshore Oil Corporation	Submarine reservoir in the Pearl River Mouth Basin, South China Sea	300000 tons/a
Jilin Oilfield CO_2 - EOR Research and Demonstration	2021	PetroChina Jilin Oilfield Company	Jilin Oilfield	2060000 tons total
CCUS Project in Shengli Oilfield	2022	Sinopec Group Co., Ltd	Shengli Oilfield	1000000 tons/a

In line with the development trend of green energy, China has introduced a series of policy guidelines to promote carbon capture, utilization, and storage (CCUS) technology, such as the "14th Five Year Plan and 2035 Long Range Goal Outline" and the "China Carbon Dioxide Capture, Utilization, and Storage Annual Report (2021)", which clearly point out the importance of developing CCUS technology in building a green energy society in China. CCUS refers to the separation and enrichment of CO_2 generated in industrial production, energy consumption, and other activities, which is then transported to factories and other places for reuse or placed on the surface, underground, and ocean for long-term storage. At present, the main development is geological storage technology, which refers to the process of injecting CO_2 into tectonics within 800-3500 m underground to achieve long-term isolation of CO_2 from the atmosphere[7]. According to statistics, the world's CO_2 geological storage capacity is about 6-42 trillion tons, while China's CO_2 storage capacity is about (1.21-4.13) trillion tons, as shown in Table 2. It can be seen that China's CO_2 storage technology has the ability to reduce carbon emissions and has great development potential.

Tab. 3. Certain CCS/CCUS project

Area	Method	Storage capacity
Songliao Basin, Bohai Bay Basin, Ordos Basin, the Junggar Basin and other major oil fields	CO_2-EOR	5.1 billion tons
Major gas fields in Ordos Basin, Sichuan Basin, Bohai Bay Basin and Tarim Basin	CO_2-EGR	90 billion tons
	Sealed in depleted gas reservoirs	153 billion tons
Songliao Basin, Tarim Basin, Bohai Bay Basin, Subei Basin, Ordos Basin	Sealed in deep salt water layer	24200 billion tons

As early as 1994, Ohgaki and Inoue[8] proposed the idea of storing CO_2 on the seabed to reduce CO_2 in the atmosphere. However, carbon sequestration on the seabed may lead to carbon dioxide leakage on the seabed, which will lead to ocean acidification and affect the marine ecological environment. In 1996, KAZUNARI OHGAKI et al.[9] experimentally confirmed the feasibility of CO_2 replacement of CH_4 in natural gas hydrates by injecting CO_2 into a three-phase mixture system of methane, liquid, and hydrate. This marked the beginning of the research on CO_2 replacement for hydrate extraction.

3. DISTRIBUTION AND EXTRACTION OF NATURAL GAS HYDRATES

Fig. 2. is the phase equilibrium curve of natural gas hydrate. Curves A, B, C and D are respectively the hydrate critical temperature curve, formation temperature curve, hydrate critical pressure curve and formation pressure curve. Since hydrates can only exist stably below the critical temperature and above the critical pressure, that is, the shaded part in the figure, This temperature and pressure condition causes natural gas hydrates to mainly exist in some marine sediments and permafrost regions[10]. Marine gas hydrates are

mainly distributed in Norway, Canada, the Gulf of Mexico, Japan, India, the Bering Strait, the South China Sea, the East China Sea of Korea, Trinidad and Tobago and other regions. The natural gas hydrates in frozen soil are mainly distributed in Alaska, McKenzie Delta in Canada and Siberia in Russia[11].

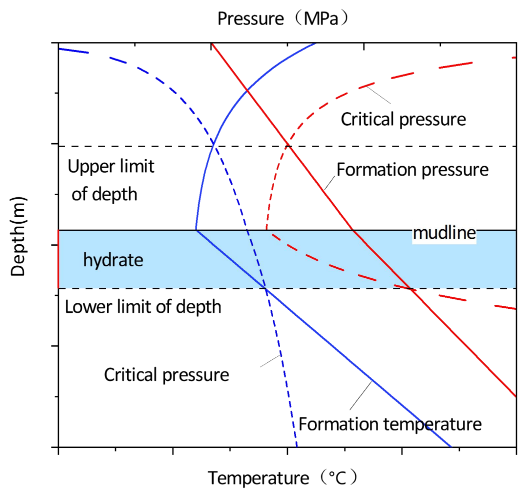

Fig. 2. Phase equilibrium Curve of Natural Gas Hydrate

Although natural gas hydrates are abundant, their sensitivity to temperature and pressure changes still poses a global challenge for commercial development. Countries such as Japan and China have actively explored this issue. Since the 1980s, Japan has been at the forefront of natural gas hydrate production: in 1995, Japan launched the CH_4 hydrate exploration and development project in the Sea of Japan; In 1997, Japan National Petroleum Corporation conducted the first successful production test on the coast of Aichi Prefecture, and produced about 120000 cubic meters of natural gas in six days; In 2013, Japan conducted offshore production trials of methane hydrates near the coasts of Atsumi and Shima Peninsula, producing approximately 120000 cubic meters of natural gas during a 6-day trial period. In 2017, they conducted the second offshore methane hydrate production experiment; In 2017, Japan successfully extracted natural gas from CH_4 hydrates near the coast of Shizuoka Prefecture through continuous production experiments. This is the first time a single well has been used to extract natural gas from methane hydrate for a long time, producing approximately 300000 cubic meters of natural gas in about two weeks. At present, Japan continues to study how to improve production technology and reduce costs to achieve the goal of establishing a commercial methane hydrate production industry by 2030[12]. Since the 1990s, China has started the exploration and research of marine gas hydrates[13], carried out drilling tests of natural gas hydrates in Shenhu, the Pearl River Mouth Basin and other regions in the South China Sea[14], [15] and successfully conducted two offshore production tests in 2017 and 2020, with an accumulated gas production of 861400 m^3 [16], laying a leading position in the exploration and development of natural gas hydrates in China.

According to the existing pilot experiments on natural gas hydrate exploration and development, although methane hydrate has the potential as a new energy source, multiple bottlenecks and challenges still need to be overcome before achieving commercial production. On the one hand, it is a safety issue. How to ensure the stability of the formation when extracting natural gas hydrates. Accidental release of methane during mining or transportation may lead to explosions or fires. The second aspect is environmental requirements. The production of natural gas from hydrates may cause environmental interference to the seabed and water bodies, and damage marine ecosystems. Meanwhile, methane is a potential greenhouse gas that, if leaked into the atmosphere, may exacerbate climate change. The third issue is cost. Due to the high cost of research, exploration, and development, the production cost of natural gas hydrates is currently high, and the low price of natural gas in the global market makes commercial production of hydrates economically advantageous. The fourth is the regulatory challenge. Given that the development of natural gas hydrates is a new phenomenon, the state must address complex legal issues such as maritime jurisdiction, resource ownership, and environmental protection when exploring and developing natural gas hydrates. Countries must work together to establish common regulations and standards to ensure the safe, efficient, and sustainable exploration and development of marine natural gas hydrates[17],[18],[19].

4. *FEASIBILITY OF CO_2 DISPLACEMENT EXTRACTION OF NATURAL GAS HYDRATES*

According to the phase equilibrium conditions of CH_4 hydrate and CO_2 hydrate, the phase equilibrium curve can be drawn, as shown in Figure 3. From the phase equilibrium diagram, it can be found that the phase equilibrium condition of CO_2 hydrate is lower than that of CH_4 hydrate, that is, under certain temperature conditions, the phase equilibrium pressure of CH_4 hydrate is higher than that of CO_2 hydrate; Or under certain pressure conditions, the phase equilibrium temperature of CH_4 hydrate is lower than that of CO_2 hydrate. Moreover, when CO_2 and free water form CO_2 hydrates, heat is released. The heat released by CO_2 hydrates per unit of substance formed is about 10kJ higher than the heat required for the decomposition of natural gas hydrates, indicating that under the same temperature and pressure conditions, CO_2 hydrates are more stable than CH_4 hydrates[20]. Therefore, the extraction of natural gas hydrates through CO_2 replacement can occur spontaneously.

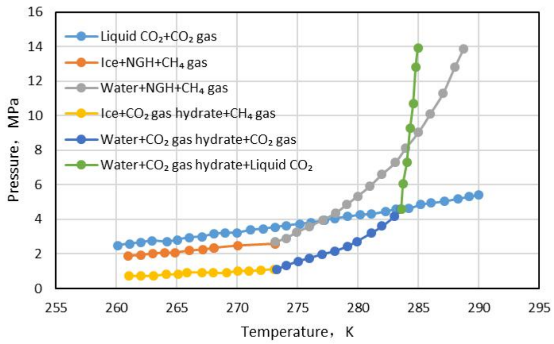

Fig. 3. The phase equilibrium curve involved in displacement

In addition, the changes in the mechanical properties of the formation after CO_2 modification of natural gas hydrate reservoirs are also an important basis for evaluating the feasibility of this technology. As early as 2013, Liu et al. conducted mechanical experiments on CO_2 hydrates, and the experimental results showed that the strength of CO_2 hydrates was higher than that of natural gas hydrates[21]. In 2015, Shen Yitao conducted compression and shear experiments on a mixture of CO_2 hydrate and natural gas hydrate, and found that the higher the CO_2 saturation, the stronger the mixture[22]. In 2018, research by Li Nan and others showed that CO_2 hydrates can improve the stability of the formation[23]. In 2022, Yan Chuanliang et al. conducted triaxial mechanical experiments on sediments containing CO_2 hydrates and natural gas hydrates, and found that the strength and inter particle cementation of CO_2 hydrates were relatively high, and the elastic modulus and strength of sediments containing CO_2 hydrates were also higher than those of sediments containing natural gas hydrates[24]. The above studies all indicate that the stability of the formation has been improved after CO_2 injection, which can meet the safety conditions for extracting hydrates. However, when CO_2 replaces CH_4, CO_2 can only occupy the space in the Type I large cage of the hydrate structure, and CH_4 cannot be displaced from the small cage. Moreover, when CO_2 and free water in the pores undergo rapid nuclear reaction, the formed CO_2 hydrate may block the pores or cover the surface of CH_4 hydrate, preventing further flow of CO_2 and reducing the replacement efficiency. Therefore, it is possible to simultaneously inject surfactants or N_2, which can dissolve or form pores on the surface of hydrates, providing a permeation channel for CO_2. N_2 can improve the fluidity of the injected fluid and fill it in small cages of hydrates, greatly improving the displacement efficiency[25],[26].

5. WELLBORE INTEGRITY CHALLENGE

The existing research focuses on how to improve the efficiency of CO_2 replacement of methane, neglecting the potential failure risks in the process and failing to provide safety assurance technologies for CO_2 replacement of methane. The process of CO_2 displacement extraction is shown in Fig. 4, which indicates that CO_2 needs to be injected into the formation containing natural gas hydrates first. CO_2 forms hydrates while releasing CH_4. There are many risk issues during the replacement process. For the downhole string of CO_2 Injection well and CH_4 production well, the corrosion of CO_2 needs to be considered, especially for the Injection well, where CO_2 is in a supercritical state when the pressure is higher than 7.2MPa and the temperature is higher than 31.3 ℃[27], the flow characteristics of supercritical CO_2 need to be considered. In addition to corrosion, it is also necessary to consider the risk of blockage after the regeneration of gas hydrates and the erosion of the inner wall of the tubing caused by the gas-liquid solid three-phase flow in the mining well. When CO_2 is injected into the formation, the high concentration of CO_2 around the Injection well may also cause hydration and chemical degradation of the cement sheath, reduce the physical and mechanical properties of the cement, lead to the formation of micro channels and micro annuli, and damage the sealing integrity of the cement. Therefore, it is necessary to study the wellbore integrity issues caused by CO_2 replacement, select safe and low-cost pipe materials and cement sheath formulas, and also optimize injection and production plans to ensure the wellbore integrity of natural gas hydrates replaced by CO_2.

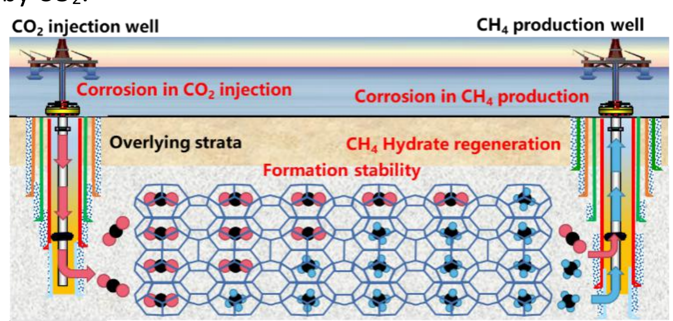

Fig. 4. CO_2 displacement extraction process for natural gas hydrates

6. CONCLUSIONS

This paper has discussed the development status and prospect of natural gas hydrate recovery by CO_2 replacement, a promising method that can achieve both energy production and carbon sequestration. It has analyzed the current situation and necessity of CO_2 storage, the distribution and challenges of natural gas hydrates in China, and the feasibility and wellbore integrity issues of CO_2 replacement extraction. The main findings are:

(1) Carbon dioxide sequestration and natural gas hydrate extraction is currently two major concerns of human society. CO_2 storage and natural gas production are both effective way to mitigate climate change and achieve China's "dual carbon goals". It is urgent to find a safe and efficient way to exploit natural gas hydrates while sealing CO_2.

(2) CO_2 replacement extraction of natural gas hydrates is a feasible method that can not only extract CH_4 but also store CO_2 in the formation. It can ensure the stability of the formation, reduce water production, lower energy consumption, and promote CCUS. However, there are still some challenges such as CO_2 injection difficulty, displacement rate limitation, efficiency improvement, and cost reduction.

(3) Wellbore integrity is an important factor that affects the safety and efficiency of CO_2 replacement extraction. It is necessary to consider the corrosion, blockage, erosion, and sealing issues of the downhole string and cement during the replacement process. It is also important to select suitable pipe materials and cement formulas, and optimize injection and production plans.

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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.

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