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GAS HYDRATE EXPLOITATION FROM WATER-SATURATED SEDIMENTS BY PRESSURE-RETAINING GAS INJECTION

Yi-Fei Sun, Jin-Rong Zhong, Rui Li, Guang-Jin Chen*, Chang-Yu Sun*

State Key Laboratory of Heavy Oil Processing, China University of Petroleum, Beijing 102249, CHINA

ABSTRACT

For the depressurization process of the water-saturated hydrate reservoir, the gas recovery is seriously restrained by the continuous seepage of the surrounding water, resulting in high water production. In response to this problem, the technique of pressure-retaining gas injection was used in this work to inhibit water production and enhance gas recovery. The results showed that water production was reduced by hundreds of times and gas recovery reached ~30% by PRGI.

Keywords: gas hydrate, pressure-retaining gas injection, CH4 recovery, water production.

1. INTRODUCTION

Natural gas hydrates (NGHs) is an ice-like crystalloid solid compound that formed by water and small molecules under low temperature and high pressure [1]. On account of the following advantages: wide distribution, large storage scale, high energy density and methane-based clean energy, NGHs is known as a potential alternative energy source in the future, thus the research on hydrate exploitation has become very hot. The conventional production methods include depressurization, thermal stimulation, inhibitor injection and CO_2-CH_4 replacement. As a dual-role method with great application prospects, CO_2-CH_4 replacement technique has undergone significant innovation. The use of CO_2/N_2 mixtures instead of pure CO_2 can largely solve the issue of low replacement efficiency and has been therefore widely studied [2-4]. In our previous work, a combined hydrate exploitation technology by integrating CO_2/H_2 injection and CH_4 production with in situ steam reformation of CH_4 was proposed [5]. The CO_2/H_2 continuous injection-production mode (CIPM) [6] and semi-continuous injection-production mode (SCIPM) [7] for CH_4 recovery were systematically studied in the gas-saturated hydrate sediments. The results show that the desirable CH_4 recovery and CO_2 sequestration can be achieved by controlling injection rate, gas composition and injection-production mode comprehensively. The optimization programs can be of significance for guiding future NGHs exploitation. However, all the previous studies have focused on gas-saturated systems, ignoring the effects of large amounts of seawater in seabed sediments.

In order to enhance the authenticity of production process and verify the applicability of the CO_2/H_2-CH_4 replacement in water-saturated sediments, in this work, we proposed a so-called pressure-retaining gas injection mode (PRGI). During the gas production process, the constant reservoir pressure is expected to restrict the flow of surrounding water, thus enormously reducing water production. This research continues to fill in the blank of the injection-production technology, and will greatly promote the practical application in the future.

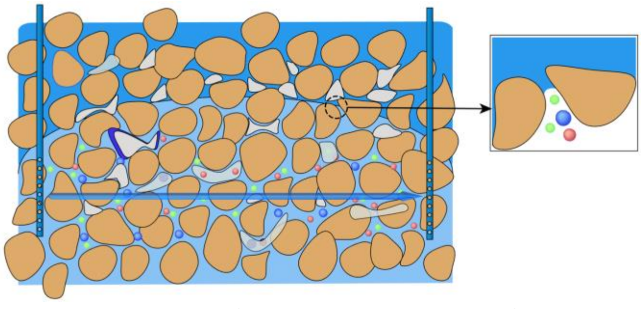

Fig. 1. Schematic of hydrate exploitation from water-saturated sediments by pressure-retaining gas injection mode.

2. EXPERIMENTAL SECTION

2.1 *Materials and experimental apparatus*

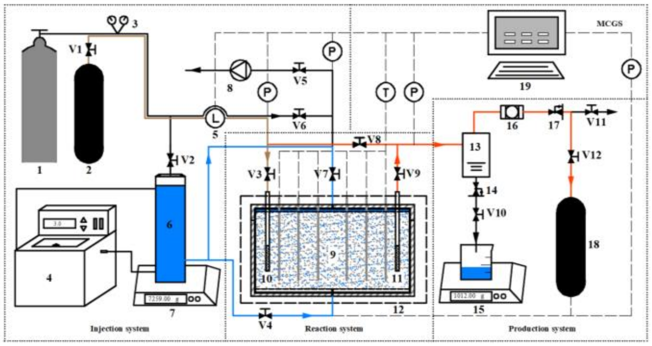

Fig. 2. Schematic graph of experimental apparatus. Injection system includes: 1- CH_4 cylinder, 2- injection gas cylinder, 3- relief valve, 4- circulator, 5- mass flow transducer, 6- water tank, 7- mass balance, 8- vacuum pump. Reaction system includes: 9- reactor, 10- injection well, 11- production well, 12- water bath. Production system includes: 13- gas-water separator, 14/17- back pressure valve, 15- mass balance, 16- filter, 18- production gas cylinder. MCGS includes all the transducers and computer.

CH_4 gas with a mole fraction purity of 99.99% and CO_2/H_2 gas mixture were both supplied by the Beijing Haipu Gas Company. LTD. The mole ratio of the binary CO_2/H_2 gas mixture determined by gas chromatography (Agilent 7890B, USA) was 56:44. Sodium sulphate solution with salinity of 33.5 g/L was prepared in the laboratory. The deionized water used was produced by a water distillation unit from Shanghai Yarong Biochemistry Instrument Factory. Sodium sulfate anhydrous with a purity of 99.5% was purchased from Tianjin Guangfu Technology Development Company. The hydrate-bearing sediment sample was simulated with 20-40 mesh quartz sands.

Fig.2 shows the schematic of experimental apparatus used in this work, which has been reported in our previous studies [6, 7]. The difference is that a constant pressure thermostatic piston tank is added to the injection system to simulate the peripheral water environment. The water in the piston tank can be pushed into the reactor when the reservoir pressure decreases.

2.2 *Experimental procedure*

The preparation of hydrate-bearing sediments in this work was the same as that used in our previous studies [6, 7]. After that, 2300g brine at 3℃ was slowly pressed into the reactor at a rate of 0.5 g/s from the bottom of the reactor, while CH_4 is continuously discharged from the top of the reactor to maintain system pressure constant. During the process, the CH_4 hydrate saturation in the sediment was almost constant, while the water saturation was greatly increased. The final properties of the hydrate sediment samples are listed in Table 1.

Table 1. Properties of prepared methane hydrate-bearing sediments and experimental conditions.

Runs	1	2	3	4
Overlying aquifer volume (L)	/	16	/	16
Temperature [a] (℃)	3.05	3.01	3.03	2.97
Pressure [b] (MPa)	3.70	3.79	3.73	3.37
Hydrate saturation (%)	23.1	23.4	22.8	24.5
Water saturation (%)	22.8	68.5	23.1	66.9
Gas saturation (%)	54.1	8.1	54.1	8.6
Production method [c]	DP	DP	GI	GI
Gas injection rate (SLM)	/	/	3.0	3.0
Gas composition (CO_2:H_2)	/	/	56:44	56:44
Production pressure (MPa)	2.0	2.0	3.6	3.6

[a] Average temperature after hydrate formation was complete.

[b] Average value of P2 and P3 after hydrate formation was complete.

[c] Gas injection (GI), Depressurization (DP).

[d] Production pressure was 2 MPa during the depressurization stage, and 3.6 MPa during the gas injection stage.

After the preparation of hydrate sample, the depressurization or pressure-retaining gas injection process was carried out. The production pressure was set to 2 MPa during the depressurization process, while 3.7 MPa during PRGI process, which was equal to the original reservoir pressure. The gas injection rate of CO_2/H_2 gas mixture was regulated to 3 SLM by a CNC gas flowmeter. The gas compositions in the reactor, the gas pipeline and the gas cylinder were measured by a gas chromatogram (GC, Agilent 7890B) at an interval of 15-30 min. When the CH_4 mole fraction in the produced gas was <10%, gas production ended. 48 h later, the reservoir temperature was raised to 15 ℃ to decompose the residual hydrates in the reactor, and before and after that the gas composition in the reactor was determined for data accounting.

2.3 *Results and discussion*

2.3.1 Temperature

As shown in Fig. 3, the temperature variations in the processes of depressurization and PRGI were quite different. Depressurization first caused a significant decrease in reservoir temperature, possibly accompanied by the formations of ice and secondary hydrates. Then the reservoir temperature gradually rose. For Run 1, the reactor was isolated, so the temperature recovery was mainly controlled by heat transfer. For Run 2, the reactor was connected with the water tank, and under the action of the pressure difference between them, the peripheral water continuously entered the sediments, resulting in a rapid increase of the reservoir temperature. However, the reservoir temperature had been stable throughout the PRGI process. The main reason was that the heat released by the formation of CO_2 hydrate compensated for that absorbed by the dissociation of CH_4 hydrate. Therefore, PRGI technology can stabilize reservoir temperature, thus avoiding the decline of gas production efficiency caused by low temperature.

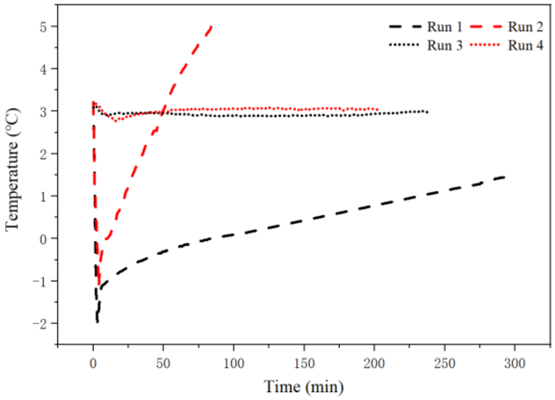

Fig.3. Variations of the average temperature for Runs 1-4.

2.3.2 CH_4 recovery

As shown in Fig. 4, there were great differences in CH_4 recovery in different occurrence environments. CH_4 recovery ratios from gas-saturated sediments by depressurization and PRGI were both much higher than that from water-saturated sediments. For the gas-saturated hydrate sediments, the effect of depressurization seemed to be better, which was due to the limited size of the simulated hydrate sample. The pressure throughout the reservoir could be rapidly reduced to the production pressure, thus stimulating the hydrate dissociation in the whole reservoir. However, when using PRGI method, the affected region by the injected gas was mainly concentrated between the injection well and the production well, while the hydrate dissociation in the edge region was controlled by the gas diffusion, relatively slow. For water-saturated hydrate sediments, the CH_4 recovery by depressurization was severely decreased to <20%. The main reason was that the external water continuously penetrates into the reservoir, which made it difficult to effectively reduce the reservoir pressure. In contrast, PRGI could slightly enhance the CH_4 recovery, but the CH_4 recovery ratio was also much lower than that of gas-saturated system. The main reason was that the high saturated water in the pores hindered the gas transfer, resulting in the contraction of the gas swept region.

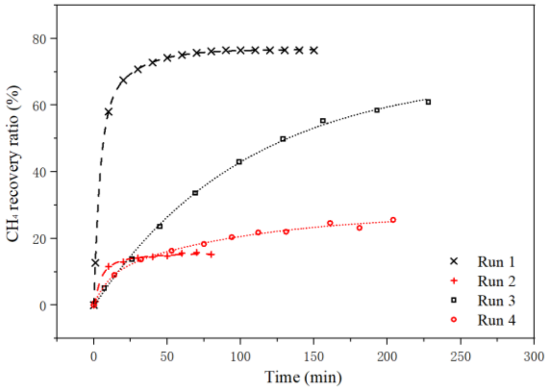

Fig. 4. Variations of CH_4 recovery ratio for Runs 1-4.

2.3.3 Water production and gas/water ratio

Fig.5 shows the accumulative water productions and gas/water ratios (R_{GW}) for Runs 1-4. Due to the low water content in the reservoir, the accumulative water productions after depressurization and PRGI were 265 and 194 g, respectively. Ultimately, the accumulative R_{GW} by PRGI was slightly higher than that by depressurization. For the water-saturated sediments, the water production and R_{GW} obtained by the two methods showed tremendous differences. Through PRGI technology, the water production decreased by ~70 times and the R_{GW} increased by ~124 times. As shown in Fig.1, the main reason for this is the formation of gas-liquid interface in the fine pore can hinder the overlying water migration. Undoubtedly, pressure-retaining operation is of significant help to the formation of such a stable gas-liquid interface.

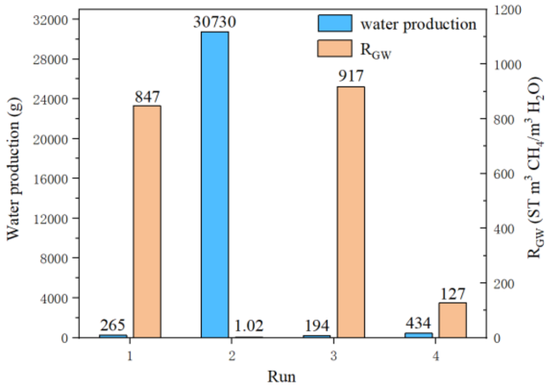

Fig. 5. Water production and R_{GW} at the end of gas production for Runs 1-4.

2.4 Conclusion

Considering the high water saturation in submarine hydrate reservoir, in this work we simulated innovative hydrate samples containing the upper constant pressure water layer and conducted a series experiments on hydrate exploitation by depressurization and PRGI. For the gas-saturated sediments, CH4 recovery by depressurization is more efficient than PRGI, but the difference in accumulative water production and R_{GW} is relatively small. For the non-enclosed water-saturated sediments, depressurization lead to the extremely low CH_4 recovery efficiency and RGW, as well as the very high water production. Exhilaratingly, the liquid migration can be significantly curbed by the formation of gas-liquid interface. Through PRGI technology, the water production tremendously decreases, thus enormously enhancing produced R_{GW}. The results is of significance for promoting the future NGHs exploitation.

ACKNOWLEDGEMENT

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