Effect of Gas Exchange Interval on CH₄ Recovery Efficiency and Study of Mechanism of CH₄ Hydrate Replacement by CO₂ Mixture

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As an environment-friendly natural gas hydrate exploitation method, CO₂ replacement method can not only achieve the purpose of mining natural gas hydrate, but also store the current greenhouse gas CO₂ in the form of hydrate on the seabed, and maintain the stratum stability of hydrate deposit area. In order to improve the rate and efficiency of CH₄-CO₂ replacement reaction, researchers proposed to use CO₂ contained gas mixture instead of pure CO₂ to replace CH₄ in natural gas hydrate. Based our previous work about CH₄ hydrate recovery with 40% CO₂ + 60% H₂, in this study, the effect of gas concentration in gas phase on final CH₄ recovery are investigated by implying different time interval of gas exchange operation. Experimental results show that The CH₄ recovery efficiency is 10.41 when the gas exchange is continues through the whole replacement process, and CH₄ recovery efficiency changes to 12.25, 32.24 and 28.86 when gas exchange operation is carried out every 12, 24, 36 h. Indicating that replaced CH₄ needs to be discharged in time to avoid CH₄ molecules being replaced to form hydrates again, and it is necessary to accurately control the time interval of gas exchange operation to avoid insufficient contact time between CO₂ and H₂ molecules and CH₄ hydrate, which affects the final replacement efficiency. In addition, the mechanism of CO₂ gas mixture containing small gas molecule such as H₂, N₂ are studied. The results indicate that when CO₂ containing small molecules such as H₂ and N₂ displace CH₄ hydrate, the existence of small molecules (H₂, N₂) can give rise to decompose the hydrate lattice and release CH₄ gas. If the gas molecules (CO₂, N₂, H₂, CH₄) in the gas phase have enough driving force to enter the hydrate lattice and remain stability, CH₄ hydrate will not decompose completely; If not, CH₄ hydrate will be completely decomposed.

Keywords: CH₄ hydrate, CO₂ replacement, gas exchange interval, mechanism, small molecules

INTRODUCTION

Natural gas hydrate (NGH), which is widely distributed in continental margin and permafrost, is naturally formed when excess gas and water molecules exist in high and low temperature zones (Sloan and Koh, 2007; Chong et al., 2016). The estimated worldwide NGH reserves are about 105–108 trillion cubic feet, twice the total reserves of natural gas, coal and oil resources (Kvenvolden et al., 1993; Boswell and Collett, 2011; Chong et al., 2016). The traditional scheme of recovering CH₄,
gas from reservoirs is to use the driving potential based on temperature, pressure and chemical potential difference to change the equilibrium condition of NGH reservoir and decompose NGH (Li et al., 2016), including thermal stimulation (Wang et al., 2017; Li et al., 2008; Fitzgerald and Castaldi, 2013), depressurization (Yang et al., 2012; Zhao et al., 2013) and chemical inhibitor injection (Yuan et al., 2011; Javanmardi et al., 2013). Besides these methods, CH4 recovery with CO2 injection into the NHG reserves was firstly proposed by Ohgaki et al. (1996), and it has become a promising way to exploit CH4 from NGH reserves while sequestrating CO2 at the same time (Koh et al., 2012; Lee et al., 2013; Bo et al., 2014; Cha et al., 2015; Zhang et al., 2017). In this method, the heat required for the decomposition of CH4 hydrate (54.49 kJ mol\(^{-1}\)) is provided by the heat released during the formation of CO2 hydrate (−57.98 kJ mol\(^{-1}\)) (Lee et al., 2003; Erslund, 2007; Falenty et al., 2016; Mu and Solms, 2017). Subsequently, the researchers proposed the exploitation of CH4 hydrate with CO2 containing mixture (Koh et al., 2012; Sun et al., 2019; Tupsakhare and Castaldi, 2019; Wang et al., 2017) and the combined use of the above methods (Li et al., 2011; Kou et al., 2019; Kou et al., 2020; Wan et al., 2020) But there are still many problems, such as low efficiency due to large energy loss to surrounding stratum for thermal stimulation, obstacles of front propagation resulted from hydrate regeneration for depressurization, environmental issues and low productivity for inhibitor injection, and inability of monitoring CO2 utilization for CO2-CH4 replacement. In addition, the influence of instability of NGH reserves has not been well understood, which may lead to sea sediment instability and more serious environmental problems because methane is an about 20 times more efficient greenhouse gas than CO2 (Dlugokencky et al., 2003).

The feasibility of replacing methane hydrate with CO2 has been proven previously (Ohgaki et al., 1996; Nakano et al., 1999), Uchida et al. (2010) investigated the CO2-CH4 replacement process with Raman spectroscopy, and found that methane can occupy large and small cages of sI hydrate, and CO2 often occupies large cages in the process of hydrate reformation. Ota et al. (2005a); Ota et al. (2005b) found that, CH4 hydrate was decomposed during the replacement process, and the decomposition rate of large cage in CH4 hydrate was faster than that of small cage. Lim et al. (2017) investigated the cage occupancy of CH4/N2/CO2 with different gas concentration and found that N2 and CO2 preferentially occupied small cages and large cages respectively. Sun et al. (2017) demonstrated that CO2 molecules in gas mixture control the entrance into hydrate cages. Wang et al. (2017) and Sun et al. (2018) studied the CH4 recovery by CO2/H2 gas mixture, and demonstrated that addition of H2 can improve the CH4 recovery.

During the replacement of CH4 in hydrate with CO2 or CO2 containing gas mixture, the concentration of each gas component in the gas phase changes as the replacement reaction proceeds. These concentration variations, especially for the CH4, affect the driving force of the gas molecule participating in the replacement and final replacement efficiency. Replaced CH4 molecules can form CH4 hydrate again or form CH4-CO2 mixed hydrate together with CO2 molecules, as a result, the new hydrate formed on the surface of the original CH4 hydrate becomes an obstacle to the further contact between CO2 or gas mixture containing CO2 with CH4 hydrate, this will eventually affect the replacement efficiency. Therefore, based on the experimental results of replacement of CH4 hydrate with 40% CO2 + 60% H2 mixture at 275.15 K, 4.5 and 6.0 MPa in our previous work (Ding et al., 2017; Ding et al., 2020), the effects of gas exchange every 12, 24, 36 h and continuous gas exchange (i.e., time interval of gas exchange) on the final replacement efficiency were studied.

Besides, the replacement mechanism of CH4 replacement from CH4 hydrate with CO2 + H2 gas mixture is also proposed: when H2 molecules contact with CH4 hydrate, the lattice of hydrate is disturbed and decomposed, and CH4 molecules escape out. If CO2 has enough driving force to replace CH4 and simultaneously occupy the hydrate cages, the hydrate lattice becomes stable again; If the driving force of CO2 molecule is not enough to occupy the hydrate cages, the lattice will be unstable and decompose to produce water and gas molecules. But when methane hydrate replaced by other gas mixtures containing small gas molecules, is the displacement mechanism the same as that of CO2 + H2 gas mixture? Therefore, 40% CO2 + 60% N2 and 20% CO2 + 80% N2 are used to study the replacement mechanism of CH4 hydrate with CO2 containing gas mixture.

**EXPERIMENTAL DEVICE AND METHOD**

**Experimental Apparatus and Materials**

The experimental device is composed of gas supply system, reactor for hydrate formation and decomposition, cooling water circulation system and detection system, as shown in Figure 1. The pure CH4 gas and 40% CO2 + 60% H2, 40% CO2 + 60% N2, 20% CO2 + 80% N2 mixtures used in experiments are supplied by Foshan Huate Gas Co., Ltd. The deionized water used in experiments is supplied by Nanjing ultrapure water technology Co, Ltd. The Raman spectrometer (LabRam, Jobin Yvon) uses 50 times long focusing lens, a 600 grooves/mm monochromator and a multi-channel air-cooled electrically coupled device (CCD) detector. It can release 532 nm wavelength laser Ar ion laser source as the laser emission source. The single crystal silicon standard sample with Raman band at 520.7 cm\(^{-1}\) is used to calibrate the Raman spectrometer.

The gas samples collected during the experiment were analyzed on Agilent 7890A, which is equipped with FID and TCD detector. The test method for gas samples is: the detector is heated from 298.15 to 523.15 K at a constant speed, the flow rate of combustion gas H2 is 30 ml min\(^{-1}\), the flow rate of combustion gas air is 400 ml min\(^{-1}\), and the flow rate of carrier gas helium is 250 ml min\(^{-1}\).

**Experimental Steps**

In order to compare the effects of different gas mixture on CH4 recovery efficiency, all experiments were carried out at 275.15 K and 6.0 MPa. The volume of reactor is 100 ml, and the amount of water used to form hydrate is 60 ml. The CH4 hydrate is
generated by bubbling at the bottom of the reactor under magnetic stirring, and the gas mixture is injected through the bottom of the reactor. About 120 h later, the water in reactor has completely transformed to hydrate which is confirmed by Raman spectroscopy where there is no characteristic peak of water, as the same as the method used before (Ding et al., 2017; Ding et al., 2020). What should be noted is that the experimental data is the average value of two groups of experiments, because each experiment was carried out in two parallel reactors.

Experiment 1, 2 and 3 were conducted using 40% CO2 + 60% H2 to replace CH4 hydrate. In Experiment 1, the mixture of 40% CO2 + 60% H2 was injected after the complete transformation of H2O to hydrate which was confirmed by Raman spectroscopy, and the top vent valve of the reactor was opened at the same time to exhaust slowly (0.45 ml min⁻¹ of exhaust speed) to shift the CH4 in gas phase to 40% CO2 + 60% H2. The top vent valve and the bottom inlet valve were kept open during the whole replacement process. What should be noted is that the pressure in reactor was remained at 6.0 MPa.

In Experiment 2, when the concentration of CH4 in the gas phase was lower than 2% during gas exchange operation, the top vent valve of the reactor was shut off, and the time marked as the beginning of the replacement reaction. After the replacement reaction proceeded 12 h, one gas sample was collected, and then the gas exchange operation was carried out. Till that CH4 concentration was lower than 2% again, another gas sample was collected as beginning of next 12 h of replacement reaction, and the top vent valve was shut off. Afterwards, the gas exchange operation was carried out every 12 h (the inlet and vent valves were opened simultaneously to inject 40% CO2 + 60% H2 gas mixture) to renew the gas in the gas phase till CH4 concentration was lower than 2% again. The gas samples were collected at the beginning and end of the exchange process and detected by gas chromatography to determine the amount of CH4 that were replaced out from hydrate phase within 12 h. Notedly, the inlet valve at the bottom of reactor was open during the whole replacement reaction. The only one difference between Experiment 3 and Experiment 2 is that the gas exchange operation was carried out every 36 h.

In order to study the reaction mechanism of CH4 hydrate replacement by CO2 mixture containing small molecules, the replacement of CH4 hydrate by CO2/N2 mixture with different concentrations was studied at different pressure. CH4 hydrate was replaced by 40% CO2 + 60% N2 mixture at 275.15 K and 6.0 MPa in Experiment 4, 20% CO2 + 80% N2 mixture at 275.15 K and 6.0 MPa in Experiment 5, and 20% CO2 + 80% N2 mixture at 275.15 K and 8.0 MPa in Experiment 6. The comparison of experimental conditions of three experiments is also listed in Table 1. Gas samples were collected every 24 h during the replacement reaction, and the concentration changes of each gas component in the gas phase in the reactor were determined by gas chromatography. After the replacement reaction, the reactor was treated with liquid nitrogen, and then the hydrate was decomposed at room temperature. The decomposed gas was collected and each component concentration in the hydrate phase was determined by gas chromatography.

RESULTS AND DISCUSSION

In our previous work (Ding et al., 2017; Ding et al., 2020), it has been repeatedly proved that the pure CH4 hydrate formed at 275.15 K and 4.5–6.0 MPa is structure I hydrate, and the
The proportion of CH$_4$ molecules in large and small cages is about 3:1, so the structure of pure CH$_4$ hydrate formed at 275.15 K and 6.0 MPa and the proportion of CH$_4$ molecules in large and small cages are not determined in this study.

Effect of Ventilation Interval on Displacement Efficiency

In Experiment 1, the gas was continuously discharged at the rate of 0.45 ml min$^{-1}$ during the whole replacement reaction (the inlet valve of 40% CO$_2$ + 60% H$_2$ was also open and the pressure in reactor was maintained at 6.0 MPa). After the replacement reaction, the gas phase was discharged quickly and the reactor was treated with liquid nitrogen. Immediately, the hydrate phase in reactor was decomposed at room temperature, and the content of each gas component originating from hydrate decomposition was determined by gas chromatography. In Experiment 1, the composition of the final hydrate decomposition gas is 89.59% CH$_4$ and 10.41% CO$_2$, that is, the recovery rate of CH$_4$ is 10.41%.

In Experiment 2, gas samples were collected at the beginning and end of the gas exchange operation, and the gas content in each gas sample were compared to have a deeper understanding of the replacement process. The increment of CH$_4$ and the decrement of CO$_2$ in the gas samples collected during the experiment are listed in Table 2, and is plotted in Figure 2.

| Table 1 | The conditions of three different gas exchange intervals in Experiment 1, 2, 3, 4, 5 and 6. |
|---------|-----------------------------------------------|
| Experiment number | Temperature (K), pressure (MPa) | Injected gas | Time interval |
| 1 | 275.15 K, 6.0 MPa | 40% CO$_2$ + 60% H$_2$ | Continuous |
| 2 | 275.15 K, 6.0 MPa | 40% CO$_2$ + 60% H$_2$ | 12 h |
| 3 | 275.15 K, 6.0 MPa | 40% CO$_2$ + 60% H$_2$ | 36 h |
| 4 | 275.15 K, 6.0 MPa | 40% CO$_2$ + 60% N$_2$ | 24 h |
| 5 | 275.15 K, 6.0 MPa | 20% CO$_2$ + 80% N$_2$ | 24 h |
| 6 | 275.15 K, 8.0 MPa | 20% CO$_2$ + 80% N$_2$ | 24 h |

| Table 2 | The CH$_4$ increment and CO$_2$ decrement in gas samples collected every 12 h in Experiment 2. |
|---------|-----------------------------------------------|
| Time item | 12 h | 24 h | 36 h | 48 h | 60 h | 72 h | 84 h | 96 h | 108 h | 120 h |
| CH$_4$ Increment (%) | 6.51 | 5.37 | 3.71 | 2.30 | 1.27 | 1.33 | 1.15 | 0.82 | 0.49 | 0.51 |
| CO$_2$ Decrement (%) | 7.66 | 6.83 | 4.46 | 3.91 | 1.57 | 1.72 | 1.67 | 1.22 | 0.71 | 0.60 |

| Table 3 | The CH$_4$ increment and CO$_2$ decrement in gas samples collected every 36 h in Experiment 3. |
|---------|-----------------------------------------------|
| Time Item | 36 h | 72 h | 108 h | 144 h | 180 h | 196 h |
| CH$_4$ increment (%) | 13.74 | 10.81 | 7.38 | 4.26 | 1.87 | 0.77 |
| CO$_2$ Decrement (%) | 15.32 | 11.45 | 7.78 | 5.11 | 2.06 | 0.99 |

FIGURE 2 | The curve of CH$_4$ increment and CO$_2$ decrement in gas samples collected every 12 h in Experiment 2.
It can be seen that the increment of CH$_4$ and the decrement of CO$_2$ in the gas samples collected every 12 h are gradually decreasing, and the decrement of CO$_2$ every 12 h is slightly higher than the increment of CH$_4$, indicating that more CO$_2$ is consumed due to the decomposition of hydrate in the replacement process. This result is consistent with the experimental results of CH$_4$ hydrate replacement with 40% CO$_2$ + 60% H$_2$ mixture, which are present in our previous work (Ding et al., 2017; Ding et al., 2020). The composition of final hydrate decomposition gas is 87.75% CH$_4$ and 12.25% CO$_2$, that is, the CH$_4$ recovery rate is 12.25%.

In Experiment 3, gas samples were collected at the beginning and end of the gas exchange operation every 36 h, the increment of CH$_4$ and the decrement of CO$_2$ in the gas samples collected during the experiment are listed in Table 3, and is plotted in Figure 3. It can be seen from the figure that the increment of CH$_4$ and the decrement of CO$_2$ decrease almost linearly, and the decrement of CO$_2$ is also slightly higher than the increment of CH$_4$ which is agreement with Experiment 2. The composition of the final hydrate decomposition gas is 72.64% CH$_4$ and 28.36% CO$_2$, that is, the CH$_4$ recovery rate is 28.36%.

The results of Experiment 1, 2 and 3 and experimental results in our previous work (Ding et al., 2020) signed as EP1, are compared in Table 4. It can be seen that different time intervals of gas exchange operation eventually led to different CH$_4$ recovery efficiency in replacement reaction. The lowest CH$_4$ recovery efficiency (10.41%) is obtained with continuous gas exchange meaning that the condition of continuous gas exchange, i.e., the mixture of 40% CO$_2$ + 60% H$_2$ passes through the hydrate area at a relatively faster speed, result in a shorter contact time between CO$_2$ or H$_2$ molecules and CH$_4$ hydrate. Thus, the replacement reaction was cannot effectively carried out, resulting in the final lower CH$_4$ recovery efficiency. The CH$_4$ recovery efficiency increased (12.25%) with the time interval changed to 12 h, and was significantly improved (32.24%) as the gas exchange interval increase to 24 h. However, when time interval increased to 36 h, the recovery efficiency decreased slightly (28.86%). The reason may be that as the time interval increases, CH$_4$ gas that replaced from hydrate phase during the replacement reaction reformed CH$_4$ hydrate again or formed CH$_4$-CO$_2$ mixed hydrate together with CO$_2$.

With the above experimental results, it can be proposed that in the real process of using gas replacement method to exploit NGH, the replaced CH$_4$ needs to be discharged from the sediment in time to avoid the replaced CH$_4$ gas forming CH$_4$ hydrate again or forming CH$_4$-CO$_2$ mixed hydrate together with CO$_2$ and so affecting the exploitation efficiency. At the same time, it is necessary to control the frequency of gas extraction from hydrate sediment, so as to avoid the incomplete contact between CO$_2$ molecules or other small gas molecules and CH$_4$ hydrate, which makes the lower CH$_4$ recovery efficiency.

The Replacement Mechanism of CH$_4$ Hydrate by CO$_2$ Mixture Containing N$_2$, H$_2$

In Experiments 4, 5 and 6, CH$_4$ hydrate was replaced by 40% CO$_2$ + 60% N$_2$ at 6.0 MPa, and by 20% CO$_2$ + 80% N$_2$ at 6.0 and 8.0 MPa. Combined with the experimental results of replacing CH$_4$ hydrate again or formed CH$_4$-CO$_2$ mixed hydrate together with CO$_2$. With the above experimental results, it can be proposed that in the real process of using gas replacement method to exploit NGH, the replaced CH$_4$ needs to be discharged from the sediment in time to avoid the replaced CH$_4$ gas forming CH$_4$ hydrate again or forming CH$_4$-CO$_2$ mixed hydrate together with CO$_2$ and so affecting the exploitation efficiency. At the same time, it is necessary to control the frequency of gas extraction from hydrate sediment, so as to avoid the incomplete contact between CO$_2$ molecules or other small gas molecules and CH$_4$ hydrate, which makes the lower CH$_4$ recovery efficiency.
CH₄ hydrate with 40% CO₂ + 60% H₂, the replacement mechanism of CH₄ hydrate by CO₂ mixture containing H₂ or N₂ was discussed.

In Experiment 4, after the water in the reactor was completely converted into hydrate, the gas mixture of 40% CO₂ + 60% N₂ was injected into the reactor until the CH₄ concentration in the gas phase was less than 2%, the replacement reaction began. The gas exchange operation was carried out every 24 h. Table 5 shows the CH₄ and CO₂ content changes in the gas samples collected during the replacement process of Experiment 4, and these data are plotted in Figure 4. It can be seen that more CH₄ is replaced in the first 48 h of the replacement process (the increments of CH₄ concentration in the gas sample every 24 h were 8 and 4% respectively), while less CH₄ is replaced out in the subsequent replacement process. This result is quite different from that of CH₄ hydrate replacement with CO₂/H₂ mixture (more CH₄ is replaced in the first 5 days). Similar to that of CH₄ hydrate replacement with CO₂/H₂ mixture, the reduction of CO₂ in gas samples is slightly higher than the increment of CH₄ every 24 h. It is suggested that the decomposition of CH₄ hydrate may also occur in the process of CH₄ hydrate replacement with 40% CO₂ + 60% N₂, resulting in the decrement of CO₂ being higher than the increment of CH₄, as observed in CH₄ hydrate replacement with 40% CO₂ + 60% H₂.

Finally, the concentration of each gas component in the hydrate decomposition gas was detected by gas chromatography, and showed as 26.31% CO₂, 2.54% N₂ and 71.15% CH₄. In other words, at 275.15 K and 6.0 MPa, the CH₄ recovery efficiency is 28.85% by using 40% CO₂ + 60% N₂, which is lower than that using 40% CO₂ + 60% H₂ at the same temperature and pressure (32.24%). However, in the experiment of replacing CH₄ hydrate with 40% CO₂ + 60% H₂, there are only CO₂ and CH₄ in the final hydrate dissociation gas, and H₂ does not exist in the hydrate phase; In contrast, there is 2.5% N₂ in the final hydrate phase in the experiment of replacing CH₄ hydrate with 40% CO₂ + 60% N₂, indicating that N₂ molecules entered the hydrate lattice and occupied the hydrate cages.

The obtained low CH₄ recovery efficiency may be resulted from that the partial pressure of CO₂ reaches 2.4 MPa (above the pressure of CO₂ hydrate formation at 275.15 K) during the replacement process, which bring about CO₂ hydrate formed quickly on the surface of CH₄ hydrate and hindered the further contact between the injected gas mixture and CH₄ hydrate, resulting in the low final replacement efficiency. So, in the following Experiment 5, CH₄ hydrate was replaced by 20% CO₂ + 80% N₂ at 275.15 K and 6.0 MPa, and the partial pressure of CO₂ was only 1.2 MPa (below the pressure of CO₂ hydrate formation at 275.15 K).

After the water in the reactor is completely converted into hydrate, 20% CO₂ + 80% N₂ is injected and the CH₄ gas in the gas phase area of the reactor is discharged at the same time. The photos of the reactor taken during the experiment are shown in Figure 5. Figure a is the picture of the reactor before injecting
20% CO2 + 80% N2 mixture, and figure b, c, d, e and f are the picture of the reactor at 4, 6, 8, 10 and 12 h after injecting gas respectively. It can be seen that most of the hydrate decomposes within 12 h, and the hydrate completely decomposes within 24 h. It shows that the mixture of 20% CO2 + 80% N2 cannot react with the decomposed water to form stable hydrate at this temperature and pressure (275.15 K, 6.0 MPa).

On the basis of Experiment 5, the reaction of CH4 hydrate with 20% CO2 + 80% N2 mixture at 275.15 K and 8.0 MPa, where the partial pressure of CO2 increased to 1.6 MPa, was carried out in Experiment 6. The experimental results are the same as those of Experiment 5. CH4 hydrate is decomposed in 24 h and no new hydrate is formed in the reactor.

In addition to the above experiments, the interaction between CH4 hydrate and pure N2 was also carried out. The pictures taken during the experiment are shown in Figure 6, the pictures a, b, c, d and e in the figure are the pictures of the reactor taken before the start of reaction and after 4, 8, 12 and 20 h of reaction respectively. It can be seen that CH4 hydrate gradually decomposes after the replacement process begins, and almost decomposes within 24 h. It shows that the contact of N2 with CH4 hydrate will lead to the destruction of hydrate lattice and release CH4 gas.

Through the above experiments, it can be seen that CH4 hydrate will be completely decomposed when the partial pressure of CO2 in the mixture is too small to form hydrate; When the mixture can form hydrate stably, CH4 hydrate will not decompose completely.

CONCLUSION

According to Experiments 1, 2, 3 and previous work, in the process of replacing CH4 hydrate with CO2/H2 mixture, the replaced CH4 needs to be discharged in time to avoid the replacement of CH4 molecules to form hydrate again. At the same time, the time interval of CH4 gas exchange process needs to be controlled accurately to avoid that the contact time of CO2, N2, H2 molecules with CH4 hydrate is not enough which affect the final replacement efficiency.

Probably, when CO2 mixture containing small molecules such as H2 and N2 replace CH4 hydrate, small molecules such as H2 and N2 attack the hydrate lattice, which can give rise to decompose the hydrate lattice and release CH4 gas. If the gas molecules (CO2, N2, H2, CH4) in the gas phase have enough driving force to enter the hydrate lattice and remain stability, CH4 hydrate will not decompose completely; If CO2 in the mixture does not have enough driving force to form hydrate or mixed hydrate, CH4 hydrate will be completely decomposed. Nevertheless, the further research is needed to elaborate the mechanism more thoroughly.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

Y-LD did the initial experiment and wrote the first draft. H-QW assisted in the experiment and revised the paper. TL proofread the figures and tables.

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