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# Experimental simulation on different formation modes of marine methane hydrate reservoirs

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# **ABSTRACT**

Marine hydrate reservoir has great resource potential, but its accumulation mode is not clear yet. At present, the study of hydrate accumulation mode is mainly carried out by in-situ exploration results and the analysis of hydrate formation conditions. In this study, methane hydrate is generated in the sediments of South China Sea on four modes, and the influence of the initial state and generation mode on the reservoir temperature and pressure change trend and the final temperature and pressure state is obtained, which provides guidance for the exploration of hydrate accumulation mode in South China Sea.

#### 1. INTRODUCTION

The global gas hydrate reserves are about  $1.5 \times 10^{16} \mathrm{m}^{3(1)}$ , which can be used as a clean energy supplement and substitute for oil and coal and has great development prospects. In China, gas hydrates are mainly distributed in the northern continental slope of South China Sea, Nansha Trough, continental slope of East China Sea and other sea areas, as well as in the Frozen regions of the Qinghai-Tibet Plateau and Mohe in northeast China. South China Sea has good conditions for gas hydrate mineralization (2). In 2017, the total methane production reached  $21 \times 10^4 \mathrm{m}^3$  in the Shenhu area of the South China Sea. And, in 2019, the second trial production of natural gas hydrate in Shenhu area in South China Sea reached  $86.14 \times 10^4 \mathrm{m}^{3(3)}$ .

In recent years, researchers have made progress on the genesis of hydrate in the South China Sea<sup>(4)</sup>. Wang et al. showed that methane hydrate could exist only in the

thermodynamically stable region with BSR occurrence on seismic plane and temperature pressure<sup>(5)</sup>. Experimental

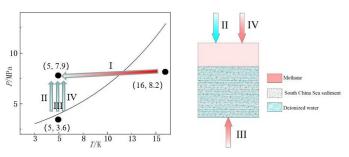


Figure 2. Sediment in the Reactor

studies show that reservoir temperature and pressure have effect on hydrate formation in sediments of the South China Sea<sup>(6)</sup>. The researchers proposed three genetic types of hydrate in the South China Sea, biogenic gas accumulated in place by way of self-generation and self-bearing, thermal gas accumulated offsite through faults and fractures by way of lower-generation and upper storage, and thermal gas accumulated offsite through mud diapir and gas chimney by way of lowergeneration and upper storage<sup>(7, 8)</sup>. The researchers also proposed some reservoir types, accretionary prism, fault-folded, dirpir, slide type and basin-edge slope<sup>(9)</sup>. In recent years, the research of methane hydrate formation has made progress<sup>(10)</sup>. Experimental studies show that the driving forces, temperature gradient and cooling rate affected the formation of methane hydrate<sup>(11)</sup>. There are few experimental studies on the origin of in-situ hydrate

in the South China Sea<sup>(12)</sup>.

On account of this, this paper, based on experimental research, systematically analyzes the process of hydrate formation on different modes,



- a. Phase equilibrium diagram
- b. Experimental schematic diagram

Figure 3. Reactor structure chart

providing reference value for further exploration of gas hydrate in the South China Sea.

#### 2. EXPERIMENTAL SECTION AND RESULT

# 2.1 Apparatus and materials

An experimental system for hydrate formation was adopted in this experiment, as shown in Figure 1. The specific layout size in the reactor is shown in Figure 2. This experimental system is mainly composed of a reaction kettle, high-precision plunger pump (ISCO pump), back pressure valve, thermocouple, water bath, gas-liquid separator, gas collector and data acquisition system. There is a water bath interlayer outside the reactor, and the top cover of the reaction kettle is fixed with the kettle body by bolts and sealed by a sealing ring. The thermocouple distribution and the distance between temperature measuring points in the reactor are shown in Figure 1.

There are 9 temperature measuring points in total for

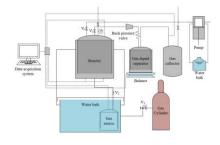


Figure 1. Experimental setup

2 vertical thermocouples. T1 thermocouple temperature measuring points from top to bottom are T11, T12, T13, T14,

T15 and T16 respectively, with the same distance between them, and T2 thermocouple temperature measuring points from top to bottom are T21, T22 and T23 respectively, with the same distance between them.

#### 2.2 Procedures and methods

This paper studies four modes to generate hydrate, and the following four methods are introduced.

All four methods of formation have the same premise: Firstly, fill 600g south China Sea sediment (110°C, drying for 24 hours) into the reactor, and then the reactor was cooled to 3°C; After the temperature stabilized, inject methane into the reactor to 2.5 MPa; Inject 250mL deionized water (3°C) was into the reactor at a rate of 5mL/min, and wait for the temperature in the kettle to stabilize.

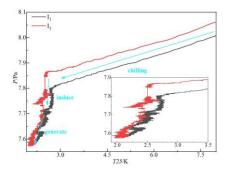
The formation steps of the first kind of mode (mode I for short) are as follows: Inject 3°C deionized water into the reactor at a rate of 50mL/min, and the pressure was 8MPa; Increase the temperature of the reactor to 16°C, keep it for 12h, drop it to 3°C, and generate hydrate. The formation steps of the second kind of mode (mode II for short) are as follows: Inject methane from top to 8MPa, and generate hydrate. The formation steps of the third kind of mode (mode III for short) are as follows: Inject methane from bottom to 8MPa, and generate hydrate. The formation steps of the third kind of mode (mode IV for short) are as follows: Inject 3°C deionized water into the reactor at a rate of 50mL/min, and generate hydrate.

The specific experimental operation parameters of this method are shown in Table 1.

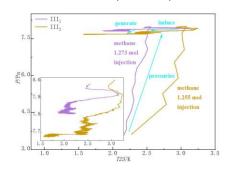
# 2.3. Generate validation

The formation of hydrate is accompanied by heat release, and the reservoir absorbs heat and the temperature rises<sup>(13)</sup>. After methane injection, the pressure and temperature of the reactor increased in this study. As the reactor is in a 3°C water bath, the heat in the reactor is taken away, and the temperature of the reactor decreases, which leads to the decrease of pressure. If hydrates are formed in the reservoir during this process, the reservoir temperature will have a special change.

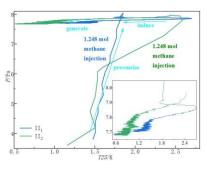
In the experiment, the reactor pressure increased from 0 to 3.45 MPa during the first injection of methane



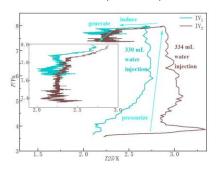
a. Pressure and temperature profiles of I



c. Pressure and temperature profiles of III



b. Pressure and temperature profiles of II



d. Pressure and temperature profiles of IV

Figure 5. Pressure and temperature profiles of four modes

and no hydrate was generated, because the temperature and pressure did not reach phase equilibrium during this process<sup>(14)</sup>. The reactor pressure increased from 4.40 MPa to 8.05 MPa during the second injection of methane, and hydrate was formed in this process. As shown in Figure 4, the dotted line represents no hydrate formation, while the solid line represents hydrate formation. In the case of no hydrate formation, each measuring point temperature will drop, and is stable at around 3°C.

In the case of hydrate formation, some measuring

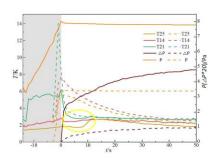


Figure 4. Temperature contrast diagram with and without hydrate formation

points temperature increase successively, and then tends to be stable. Final temperature range of hydrate formation is wider and lower than that of no hydrate formation, which reflects the particularity of hydrate formation. The measuring point T16 temperature does not increase obviously with other measurement points,

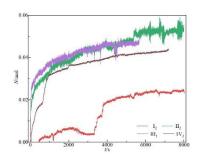


Figure 6. Profiles of methane hydrate formation process by four modes

indicating that there is almost no hydrate formation in the reservoir near this measurement point.

## 2.4. Formation features

With the process of hydrate formation, the temperature and pressure of the reservoir tend to decrease. Figure 5 showed the variation trend of pressure with temperature in the four kinds of mode to form hydrate reservoir. The decreasing trend of temperature and pressure is the same in a formation mode, and the decreasing trend of temperature and pressure is different among different formation modes.

The generation process of four modes has pressurize, induce and generation three stages, which are highlighted in figure 5. The amount of secondary injection of methane/deionized water is also indicated. Cooling the reservoir to reach above the equilibrium of

hydrate phase and generate hydrate is used in Mode I. During hydrate formation, the temperature and pressure of the reservoir has been decreasing. Injecting methane from top to increase reservoir pressure and generate hydrate is used in Mode II. The two groups showed different trend in the early part of the experiment, which may be caused by the difference in initial temperature. The subsequent processes follow the same trend. Injecting methane from bottom to increase reservoir pressure and generate hydrate is used in Mode III. Injecting deionized water from top to increase reservoir pressure and generate hydrate is used in Mode IV.

During the whole process of hydrate formation, mode I and mode II take a longer time to generate than mode III and mode IV. A higher initial temperature leads to a lower final pressure, which shown in Figure b, Figure c and Figure d. The variation of hydrate generation with time in the four ways is shown in Figure 6.

# 2.5 Conclusion

Four kind modes of increasing pressure, decreasing temperature or supplementing gas/water to form hydrate reservior were studied in this paper. The different operation modes show that both the formation mode and the initial state of reservoir will affect the formation process of reservoir hydrate, in which the formation mode plays a decisive role, and the initial state of reservoir mainly affects the final state of hydrate formation. These results are of great significance to understand the formation mechanism of methane hydrate reservoirs in nature.

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