# EXPERIMENTAL STUDY OF METHANE HYDRATE DISSOCIATION BEHAVIOR UNDER DIFFERENT PRODUCTION PRESSURE

Xiao-Yan Li<sup>a,b,c,d,e</sup>, Yi Wang<sup>a,b,c,d</sup>, Xiao-Sen Li<sup>a,b,c,d\*</sup>

<sup>a</sup> Guangzhou Institute of Energy Conversion, Chinese Academy of Sciences, Guangzhou 510640, China
<sup>b</sup> CAS Key Laboratory of Gas Hydrate, Guangzhou 510640, China

<sup>c</sup> Guangdong Provincial Key Laboratory of New and Renewable Energy Research and Development, Guangzhou 510640, China <sup>d</sup> Guangzhou Center for Gas Hydrate Research, Chinese Academy of Sciences, Guangzhou 510640, China

<sup>e</sup> University of Chinese Academy of Sciences, Beijing 100049, China

## ABSTRACT

As we all know, depressurization is considered as the most promising method to release natural gas from hydrate reservoirs. In this study, we study methane hydrate dissociation behavior in porous media under different production pressure. The experimental results show that the process of methane hydrate dissociation can be divided into the depressurization stage (DS) and the constant pressure stage (CPS). In DS, the heat required for hydrate dissociation is from the sensible heat of hydrate-bearing sediments. As the production pressure decreases, the total amount of gas production and water production increase. In CPS, the required heat for hydrate dissociation is transferred from surroundings. With the decrease of the production pressure, the rising rate of average temperature in hydrate-bearing sediments increases, and the gas production rate increases.

**Keywords:** methane hydrates, dissociation behavior, production pressure, gas production

# 1. INTRODUCTION

Natural gas hydrates are solid crystalline compounds, which are formed by water and gas molecules (such as methane) under low temperature and high-pressure conditions [1]. There are four main production methods for gas hydrate: depressurization method [2], heat stimulation method [3], the use of inhibitors [4] and the displacement method [5]. They can be used singly or in combination [6].

Depressurization is considered as the most promising method due to the low energy input [7]. Yousif et al [8] found that the gas production was determined by several factors, and the production pressure was a key factor to control hydrate dissociation. Ji et al.[9] found that the gas production rate was a sensitive function of the well pressure, and the motion rate of the dissociation front decreased with the increase of the well pressure. Hong et al.[10] observed that the over-low wellbore pressure could result in the reservoirs temperature falling below the freezing point and cause the ice formation, which is disadvantage for gas production from reservoirs. The effects of production pressure on hydrate dissociation have been studied by some researchers. However, the experimental methods are less, especially for methane hydrate dissociation in porous media.

The objectives of this work are to study methane hydrate dissociation behavior in porous media under different production pressure and to discuss the effects of production pressure on the temperature and gas production.

# 2. EXPERIMENTAL METHODS

# 2.1 Experimental apparatus

The experimental schematic diagram of PHS (Pilot Hydrate Simulator) is shown in Fig 1. The detail illustration can be found in previous study [11]. A brief

<sup>\*</sup> Corresponding author. Tel: +86-20-87057037; fax: +86-20-87034664.

*E-mail address:* lixs@ms.giec.ac.cn Selection and peer-review under responsibility of the scientific committee of the 11th Int. Conf. on Applied Energy (ICAE2019). Copyright © 2019 ICAE

description is given here. The apparatus is composed of five parts: the gas-water supply system, the highpressure reactor, the gas-water production system, the constant temperature system and the data collection system. The gas-water supply system is used for injection the water and methane gas into the high-pressure reactor, including an advection pump, an electrical balance, etc. The core part of PHS is the high-pressure reactor (0-30 MPa). The temperature of the reactor is controlled by a cold room and a water jacket (15-30 °C, ±0.5 °C). The gas-water production system is used for controlling and measuring the dissociated gas and water from the reactor, including a back-pressure valve, a gas flow meter, an electrical balance, etc. All experimental data are recorded on computer through the data collection system.



Fig 1 Schematic diagram of PHS

The distribution of production well in the highpressure reactor is shown in Fig 2. The interior of the reactor is divided into: A-A layer, B-B layer, C-C layer. In the upper flange, there are 9 vertical well positions, namely V1, V2, ...V9. V5 (production well) is located at the center of the reactor. There are 4 symmetrical grooves distributed in the production well. The distribution of thermal couples in the reactor is similar with the production well distribution. The total number of thermal couples in the reactor is 147, and T25 is located at the center of the reactor.



Fig 2 Well design and thermal couple distribution

## 2.2 Experimental procedures

The process of methane hydrate formation and dissociation in porous media has been comprehensively illustrated by Wang [12]. Therefore, a brief description of the experimental process is given here. The quartz sand with diameters of 250-425 µm was firstly packed into the reactor, and the reactor was evacuated by a vacuum pump. Then, the deionized water was added to the reactor. Next, methane gas was pressurized into the reactor to 20 MPa, and the reactor was kept at environment temperature for more than 4 fours to ensure no leakage. After that, the temperature was gradually lowered to the experimental temperature 7.0 °C. When the temperature was lowered to the equilibrium temperature, methane hydrate began to form in the reactor. The process of methane hydrate formation was considered being completed when the pressure decreased to 11 MPa.

After the formation process being completed. The outlet pressure was gradually decreased to the production pressure by controlling the back-pressure valve. A total of three experiments for methane hydrate formation and dissociation were conducted, and the production pressure were 4.7MPa, 4.2MPa, 3.7Mpa, respectively.

## 3. RSEULTS AND DISCUSSIONS

#### 3.1 The pressure profiles



Fig 3 Time dependence of pressure

Fig 3 shows the time dependence of pressure in the reactor under different production pressure. It can be known that the duration of the whole experiments decreases with the decrease of the production pressure. From the picture, the whole experimental process can be divided into two stages. The one is depressurization stage (DS): the pressure in the reactor decreases from the initial pressure to the production pressure. The

another one is constant pressure stage (CPS): the pressure keeps at the production pressure.

## 3.2 Average temperature profiles

Fig 4 shows the time dependence of average temperature in the reactor under different production pressure. The average temperature is defined as the average value of T1A~T49A, T1B~T49B, T1C~T49C.



Fig 4 Temperature profiles for different experiments

In DS, although the pressure is gradually decreased, the temperature is almost not changed since the pressure is higher than the equilibrium pressure. When pressure is decreased to below the equilibrium pressure, due to the sensible heat consumed for the endothermic reaction of hydrate dissociation, the average temperature is quickly decreased to the lowest which is the equilibrium temperature at the production pressure. From the picture, it can be known that the lowest temperature of hydrate-bearing sediments decreases with the decrease of the production pressure. There would be the risks of ice formation in hydrate-bearing sediments if the production pressure is too low. The ice formation would cause the blockage of the gas flow channel, which is disadvantage for gas production from hydrate-bearing sediments.

In CPS, as the heat gradually transfers from the surroundings to the reactor, the average temperature gradually increases from the lowest to the environmental temperature. For different experiments, the rising rate of temperature is different. The rising rate is largest when the production pressure is 3.7 MPa and is smallest when the production pressure is 4.7 MPa. The reason is that the temperature difference between the surroundings and hydrate-bearing sediments is largest when the production pressure is 3.7 MPa. The reason is that the temperature difference between the surroundings and hydrate-bearing sediments is largest when the production pressure is 3.7 MPa. Therefore, the heat driving force is largest, and the rising rate of temperature is largest. It can be concluded that the rising

rate of temperature during hydrate dissociation increases with the decrease of the production pressure.

## 3.3 Gas production

The time dependence of gas production for different experiments are shown in Fig 5. From the picture, it can be known that the total volume of gas production is almost same, since the initial conditions is consistent. In DS, the total gas volume produced increases with the decrease of the production pressure. The reason is that the required heat for hydrate dissociation in DS is provided by the sensible heat of hydrate-bearing sediments, and the magnitude of sensible heat determines the amount of hydrate dissociation. The sensible heat is caused by the difference between the initial temperature and the equilibrium temperature at the production pressure. As the production pressure decreases, the sensible heat of hydrate-bearing sediments increases. Therefore, the total volume of gas produced in DS increases with the decrease of the production pressure.



Fig 5 Time dependence of gas production

In CPS, the highest gas production rate (the slope of the curve) is obtained when the production pressure is 3.7 MPa, and the lowest gas production rate is obtained when the production pressure is 4.7 MPa, and the gas production rate of 4.2 MPa is between them. The reason is that the required heat for methane hydrate dissociation in CPS is transferred from the surroundings. Therefore, methane hydrate dissociation rate is controlled by the heat transfer rate from the surroundings to the reactor. As we discussed above, the heat transfer rate increases with the decrease of production pressure. The heat transfer rate is largest when the production pressure is 3.7 MPa. Therefore, methane hydrate dissociation rate is fastest, and the gas production rate is largest.

## 3.4 Water production

Fig 6 shows the time dependence of water production. It can be known that the total amount of water production increases with the decrease of the production pressure. Since the flow driving force  $\Delta P$  (the difference between the production pressure and the initial pressure) increases with the decreases of the production pressure. For different production pressure, the water produced in DS is larger than that in CPS. The reason is that the hydrate-bearing sediments is water-saturated. In DS, the pore in the sediments is filled with water. Therefore, the amount of water production is large. In CPS, the pore is gradually filled with gas due to hydrate dissociation, and water is discontinuous. Therefore, the amount of water production gradually decreases.



Fig 6 Time dependence of water production

## 4. CONCLUSIONS

In this study, methane hydrate dissociation behavior under different production pressure is studied. There are some main conclusions: (1) in DS, as the production pressure decreases, the lowest temperature of hydratebearing sediments decreases, and the total amount of gas production and water production increase. (2) in CPS, with the decrease of the production pressure, the rising rate of average temperature in hydrate-bearing sediments increases, and methane hydrate dissociation rate increases. In actual field, it's beneficial to decrease the production pressure, since it can improve the gas production rate. However, the production pressure shouldn't be over-low, since there could be the risks of hydrate reformation which is disadvantage for gas production from hydrate reservoirs.

## ACKNOWLEDGEMENT

This work is supported by Key Program of National Natural Science Foundation of China (51736009), National Natural Science Foundation of China (51676190), Pearl River S&T Nova Program of Guangzhou (201610010164), International S&T Cooperation Program of China (2015DFA61790), which are gratefully acknowledged.

## REFERENCE

- [1].Sloan, E.D. and C.A. Koh, Clathrate Hydrates of Natural Gases, Third Edition. 2008.
- [2].Yousif, M.H., et al., Depressurization of Natural-Gas Hydrates in Berea Sandstone Cores. Journal of Inclusion Phenomena and Molecular Recognition in Chemistry, 1990. 8(1-2): p. 71-88.
- [3].Tang, L.G., et al., Experimental investigation of production behavior of gas hydrate under thermal stimulation in unconsolidated sediment. Energy & Fuels, 2005. 19(6): p. 2402-2407.
- [4].Li, G., et al., Experimental investigation of production behavior of methane hydrate under ethylene glycol injection in unconsolidated sediment. Energy & Fuels, 2007. 21(6): p. 3388-3393.
- [5].White, M. and P. McGrail, Designing a Pilot-Scale Experiment for the Production of Natural Gas Hydrates and Sequestration of CO(2) in Class 1 Hydrate Accumulations, in *Greenhouse Gas Control Technologies 9*. 2009. p. 3099-3106.
- [6].Song, Y.C., et al., Evaluation of gas production from methane hydrates using depressurization, thermal stimulation and combined methods. Applied Energy, 2015. 145: p. 265-277.
- [7].Li, X.S., et al., Experimental investigation into gas production from methane hydrate in sediment by depressurization in a novel pilot-scale hydrate simulator. Applied Energy, 2012. **93**: p. 722-732.
- [8].Yousif, M.H.S., E. D., Experimental Investigation of Hydrate Formation and Dissociation in Consolidated Porous Media. SPE Reservoir Engineering (Society of Petroleum Engineers), 1991. 6(4): p. 452–458.
- [9].Ji, C., G. Ahmadi, and D.H. Smith, Natural gas production from hydrate decomposition by depressurization. Chemical Engineering Science, 2001. 56(20): p. 5801-5814.
- [10].Hong, H. and M. Pooladi-Darvish, Simulation of depressurization for gas production from gas hydrate reservoirs. Journal of Canadian Petroleum Technology, 2005. 44(11): p. 39-46.
- [11].Feng, J.C., et al., Effect of horizontal and vertical well patterns on methane hydrate dissociation behaviors in pilot-scale hydrate simulator. Applied Energy, 2015. 145: p. 69-79.
- [12].Wang, Y., et al., Analytic modeling and large-scale experimental study of mass and heat transfer during hydrate dissociation in sediment with different dissociation methods. Energy, 2015. **90**: p. 1931-1948.