



## Full Length Article

# Production characteristics of two class water-excess methane hydrate deposits during depressurization

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## ABSTRACT

There are four main classes of methane hydrate deposits in nature. These classes differ in their distributions of the methane hydrate layer, free water layer and free gas layer. The gas production rate, water production rate, gas-to-water ratio, gas and water production recovery during the depressurization dissociation process are different for hydrate samples of different classes. Water-excess hydrate samples of Classes 2 and 3 were prepared in the present work, and they were dissociated at 2.0 MPa, 2.3 MPa, 2.6 MPa and 3.0 MPa. Ice formation was observed when the backpressure was 2.0 MPa. The gas-to-water ratio for Class 3 samples was higher than that of Class 2 samples, regardless of the backpressure. The average gas production rate for the 90% overall gas amount of Class 2 samples was higher than that of Class 3 samples for a backpressure of 2.0 MPa, whereas the opposite result was found for backpressures equal to or greater than 2.3 MPa. Furthermore, the water recovery value of Class 2 samples was higher than that of Class 3 samples by approximately 10%, and the gas recovery value of Class 2 samples was lower than that of Class 3 samples by approximately 20% at the same backpressure. These experiments provided some basic data for the exploitation of Class 2 and Class 3 deposits and find out the influence of backpressure on methane hydrate dissociation of the two class deposits by depressurization.

## 1. Introduction

In the Reference Case of the BP Energy Outlook, demand for natural gas will increase annually by 1.6% until 2035, exceeding the increase in demand for oil and coal. The shale gas supply is expected to increase at the fastest rate (5.2% every year) until 2035 due to the shale gas revolution in the United States [1]. Methane hydrate is also a natural gas resource with amounts that are far greater than conventional natural gas resources. If it can be exploited commercially, methane hydrate will be another natural gas supply resource, so it has been studied in many countries [2]. Methane hydrates are an ice-like crystal substance including water and methane. The water forms cage structures through hydrogen-bond interactions with methane bound in the cage as the guest molecule [3]. Methane hydrate formation requires high pressure and low temperature, and therefore methane hydrates are always present in permafrost and marine continental slopes [4]. Four main classes of methane hydrate deposits are found in nature [5]. Class 1 deposits, such as those found in the North Slope of Alaska [6], include two layers between impermeable layers, with the hydrate layer as the upper layer and a gas-liquid two phase that includes mobile free gas as the under layer. Class 2 deposits, such as those found in the Nankai Trough of Japan [7], also include two layers, and the upper layer is also a hydrate

layer; the lower layer is a mobile water layer. Class 3 deposits, such as those found in the Qilian Mountain permafrost in China, include only the methane hydrate layer between the impermeable layers. Class 4 deposits, such as those found in the Krishna Godavari basin in India [8], include a low methane hydrate saturation (< 10%) layer and no impermeable layers. Class 2 and Class 3 deposits are studied in the present work.

To compare the classes of deposit, Moridis et al. simulated Class 1, Class 2, Class 3 and Class 4 deposits and studied their gas production rates, the accumulation of gas production during the methane hydrate dissociation [6,9–13]. Konno et al. applied MH21-HYDRES to simulate the gas production of Class 1–3 marine methane hydrate deposits by depressurization. They found that some Class 2 and 3 deposits in the Gulf of Mexico and Eastern Nankai Trough (permeability: 0.001–100 mD) are suitable for depressurization because their permeability exceeded the threshold value (1–10 mD) and the temperature was high [14]. Zhao et al. applied Tough + Hydrate to simulate gas production of Class 3 deposits which are in the Qilian Mountain permafrost of Qinghai-Tibet Plateau by depressurization, and they found that single vertical well method is not ideal method for this place [15]. Bhade et al. simulated Class 2 methane hydrate deposits with a thickness of 10 m, an aqueous layer of 2 m and a length and the width of

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240 m, considering 4 production wells. They found that the hydrate dissociated earlier at the interface of two hydrate-bearing layers [8]. Xia et al. used Hydrate-Res-Sim to investigate the production characteristics for three classes of methane hydrate deposits by depressurization and thermal stimulation as well as by the combination of these two methods. They simulated a Class 2 deposit with a 15-m thick hydrate layer and 15-m thick water layer and a Class 3 deposit with a thickness of 18.25 m. They found that for Class 2 deposit hydrates, the saturation influence was greater than the effect of backpressure. For Class 3 deposit hydrates, the saturation influence was greater than the effect of reservoir temperature [16].

To date, studies of different classes of deposits have been mostly performed by simulation, and there have been few experiments on the different classes of methane hydrate deposits. However, there have been many experimental studies on the dissociation of water-excess methane hydrate deposits [17–20]. Additionally, among depressurization, heat stimulation and inhibitor injection, the depressurization method was reported to be the best approach for the exploitation of methane hydrate deposits [7,21–22]. There have been some studies on the dissociation of water-excess methane hydrate deposits by depressurization. For example, Li et al. studied low gas saturated methane hydrate deposits with a water saturation of approximately 46% to simulate marine hydrate deposits. They found that free gas saturation was an important factor influencing the production behavior [23]. Yang et al. applied magnetic resonance imaging (MRI) to study gas production from deposits at different water saturation levels by depressurization and found that an increase in water saturation leads to a decrease in the gas production rate [24]. Chong et al. studied the dissociation of water-saturated methane hydrate deposits by injecting water several times during the formation to induce homogeneous formation and discussed the influence of the backpressure on gas and water production, finding that the cumulative amount of water increased with increasing backpressure [25].

The experiments performed in the present work were also focused on the dissociation of methane hydrate deposits with excess water by depressurization. However, this study is different from previous studies. The dissociation of the two classes of methane hydrate deposits were compared for two cases. The first case (Class 3) was similar to the previous studies, focusing on the water-excess methane hydrate layer, whereas the second case (Class 2) was different from previous studies and included two layers, with the under layer full of water in sands without the gas, and the upper layer a water-excess methane hydrate layer. There was little experimental study about Class 2 samples. This study used a simple method to construct Class 2 sample, and only changed the glass beads and water filling method to construct Class 2 samples. The gas recovery, water recovery, gas and water production rates, and heat and mass transfer were compared for the two types of methane hydrate deposits. The results obtained in this work will provide fundamental experimental data for subsequent correlational research. It will be helpful for analyzing the mining of methane hydrates in real marine continental margin sediments.

## 2. Experimental material and methods

### 2.1. Experimental apparatus and materials

The experimental equipment is shown in Fig. 1. The equipment consists of four components, namely, the injection component, the reactor, the gas and water collection components and the data collection component.

A gas pump and a water pump (260D, Teledyne Isco, Inc., USA) were used to inject the gas and water into the reactor, respectively. The reactor material is stainless steel, and it can withstand pressures of 15.0 MPa. The reactor's inner height was 120 mm, and its inner diameter was 103 mm. The reactor temperature was controlled by the water bath in the 263.15–300.15 K range. There were 17 thermocouple

points in the reactor for real-time monitoring of the temperature. The distribution of the thermocouple points is shown in Fig. 1. The 17 thermocouple points were distributed in 3 thermocouples that were evenly spaced in a circle with a diameter of 60 mm, and the distance between neighboring points was 20 mm. The three thermocouples were named T1, T2 and T3. The measurement points of T1 were named T11, T12, T13, T14, T15 and T16, the measurement points of T2 were named T22, T23, T24, T25 and T26, and the measurement points of T3 were named T31, T32, T33, T34, T35 and T36 from top to the bottom. The heights of the three thermocouples were not the same; T22 was higher than T32 by 5 mm, and T32 was higher than T12 by 7 mm. The gas and water collection component included a gas-water separator and a gas collection vessel. The gas-water separator was used to collect the water and separate it from the gas, and its quality was monitored by a balance. The data collection component included the thermocouples and the pressure gauge. The precision of the thermocouples and the pressure gauge was  $\pm 0.1$  K and 0.1 MPa, respectively.

High purity methane (99.99%, Dalian Special Gases Co., LTD, China) was used. BZ 01 glass beads (produced by Japan AS-ONE Company) were packed into the reactor to simulate the porous medium, and its porosity was approximately 34.8%. Its density was  $2.5 \text{ g/cm}^3$ , and the particle size range was 105–125  $\mu\text{m}$ . The absolute permeability of the beads was  $7.8 \mu\text{m}^2$ . All experiments used deionized water.

### 2.2. Experimental method and procedure

Two filling methods were used to construct the two classes of water-saturated methane hydrate samples. The first method, consisting of an overall reactor filled with dry glass beads at first, was used to simulate Class 3 samples. The other method made the bottom 32 mm layer filled with water-saturated glass beads, the upper 88 mm filled with dry glass beads, which can simulate Class 2 samples. The bottom water-saturated beads layer included beads and a calculated amount of water that exactly make the bead pores full of water. And there was nothing between two layers for Class 2 samples. Then, the reactor was connected to the experimental system. Nitrogen (8.5 MPa) was injected to determine whether the device leaked gas. The experiment could be performed when the reactor gas pressure was constant at 8.5 MPa for 12 h and the pressure fluctuated by less than 0.1 MPa. Next, the nitrogen was released slowly to atmospheric pressure. Then, a quantity of methane was injected into the reactor using the gas pump. After the methane injection, water was first injected to reach a pressure of approximately 8.0 MPa at room temperature (23 °C). Then, the water bath was used to cool the reactor to 2.0 °C, leading to hydrate formation. Once the pressure decreased to approximately 3.2 MPa due to hydrate formation, and it can keep constant for longer than 30 min. Then, water at a temperature of 2.0 °C was second injected into the reactor to reach a pressure of approximately 8.0 MPa for the further formation of the hydrate. Then, as before, when the pressure decreased to a value and keep constant for longer than 30 min, water at a temperature of 2.0 °C can be third injected into the reactor to approximately reach a constant pressure of 8.0 MPa. When the pressure decreased slowly with time after this water injection, the 2.0 °C water was injected into the reactor a fourth time to reach a pressure of approximately 8.0 MPa. The pressure remained almost constant after the fourth water injection, and the temperature was set to 4.0 °C to make the sample more similar to real methane hydrate deposits. Chong et al. [25] and Li et al. [23] applied a similar method for sample fabrication. Methane hydrate dissociation began after sample preparation. The backpressure valve and V-2 were turned on to depressurize, which can lead to hydrate dissociation. The backpressure was set to 2.0 MPa, 2.3 MPa, 2.6 MPa and 3.0 MPa for the different classes. All backpressure values were lower than the phase equilibrium pressure (3.85 MPa) at 4.0 °C, which can be obtained according to *hydiff.exe* [26].

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