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EFFECT OF OVERLYING WATER ON THE FORMATION AND DISSOCIATION OF METHANE HYDRATES IN SEDIMENTS

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ABSTRACT

Methane hydrate (MH) is considered as one of the most promising clean energy. In this study, the effects of overlying water on the formation and dissociation of MHs in sediments were analyzed. The results indicate that the overlying water can provide the sufficient water source and the driving force for hydrate formation. The time required to synthesize hydrate sample with high saturation (44.1%) is effectively shortened. Additionally, the overlying water contributes to the heat and mass transfer, thereby promoting the hydrate dissociation and the gas-water production during depressurization. The findings of this study could pave the way for the utilization of overlying water to enhance the gas production efficiency for marine hydrate exploitation.

Keywords: methane hydrate, overlying water, dissociation, sediment, depressurization

NOMENCLATURE

Abbreviations	
MH	Methane hydrate
Symbols	
M	Molar mass
m	Weight
n	Amount of mole
P	Pressure
S	Saturation
T	Temperature
t	Time
V	Volume
v	Rate
φ	Porosity
ρ	Density

1. INTRODUCTION

Natural gas hydrates are widely distributed in permafrost regions and shallow sediments on marine continental margins [1]. Although a series of hydrate coring and field production trials have been conducted over the past two decades, commercial exploitation has proven to be challenging and has not been achieved yet.

Globally, 98% of naturally occurring MHs exist in the seabed sediments [2], where the gas and the water supply is plentiful, and the low-temperature and high-pressure conditions are available. Marine hydrates have attracted global attention as one of the most potential clean energy sources. According to the data of logging interpretation and seismic profiles, the hydrates in many marine environments are shallowly buried or even exposed on the seafloor [3, 4]. Therefore, for the field exploitation and commercial application of the gas recovery from marine hydrate reservoirs, it is necessary to consider the influence of overlying water. Related scholars have carried out some numerical simulations and experimental researches on the formation and dissociation of hydrate in sediments [5], and have made considerable progress. However, few experimental studies on the effects of overlying water on the formation and dissociation of MHs in sediments. Thus, the effects of overlying water on gas-water production from hydrate reservoirs by steady-rate depressurization remain an issue to be address.

In this study, after MHs were synthesized in sediments, steady-rate depressurization was conducted to produce gas and water from hydrate-bearing sediments in the presence of overlying water. The effects of overlying water on the formation and dissociation of MHs in sediments were analyzed.

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2. EXPERIMENTAL SECTION

2.1 Experimental apparatus

An experimental setup was built to study the effects of overlying water on the formation and dissociation of MHs in sediments, as shown in Fig. 1. The experimental apparatus includes a cylinder reactor with a volume of 0.98 L. A vertical wellbore with a borehole design at the bottom section was located at the center of the reactor.

A syringe pump (500D, Teledyne ISCO Inc., USA) is used for water injection at a constant pressure/flow mode. A T-type thermocouple (± 0.1 K) is installed at the outlet of the pump to measure the temperature of water flowing out of the pump. Depressurization simulated well is sequentially connected to a control valve (Fisher-Baumann, USA), a gas-water separator of 1.0 L placed in a precision balance (±0.1 g, KERN 572-57), and a gas collector of 4.785 L. Two T-type six-point thermocouples (± 0.1 K) are used to monitor the temperature of the entire reservoir. Two pressure transducers (±0.02 MPa) are applied to measure the pressures at the top and bottom of the reactor, while one additional pressure transducer is used to measure the pressure of gas collector. Data acquisition system integrated with LabView software was used to record the real-time experimental data.

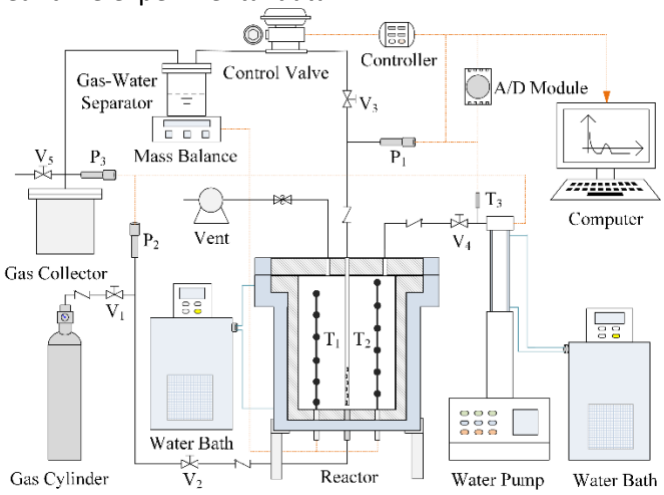

Fig. 1 Schematic diagram of the experimental system.

2.2 Materials

Methane gas of 99.99% purity was supplied by Air Liquide Singapore Pte Ltd. The unconsolidated silica sand with a particle size ranging from 0.10 to 0.60 mm (D_{50}= 0.25 mm) was supplied by River Sands Pty Ltd. The deionized water was used to create the aqueous-rich sediments environment for hydrate formation and dissociation.

2.3 Experimental procedures

First, 1480 g of dried silica sand was tightly packed in the reactor to create a porous medium with a porosity of 42.9%. After leak detection, the reactor and the connecting tubing were purged with low-pressure (~1.0 MPa) methane three times.

The subsequent experimental procedures, as well as the temperature and pressure evolution corresponding to each procedure, are shown in Fig. 2. Each step of the experimental procedure was carried out after the temperature and pressure have stabilized. Inject quantitative gas and water into the reservoir above the hydrate phase equilibrium (A-D). Then, wait for the temperature to stabilize and the water-gas phases to redistribute (D-E). In this study, the water injection pump that simulates the overlying water provides a constant pressure environment and induces hydrate formation by cooling to 274 K (E-F). When the water consumption rate is lower than 0.6 mL/hr (usually takes approximately 60 hr), the temperature of the reactor was increased to 279 K (corresponding to the temperature at ~100 m below the seafloor) (F-G).

Steady-rate depressurization was initiated by slowly opening valve V_3 and starting the program of the control valve to induce hydrate dissociation and gas-water production (G-H). In this study, the pressure drawdown rate was approximately 3.0 MPa/hr, and the exploitation pressure was 3.0 MPa. The water pump with a constant flow rate of 1.0 mL/min was always connected to the reactor for simulating the erosion of hydrate-bearing sediment reservoirs by the overlying water during depressurization.

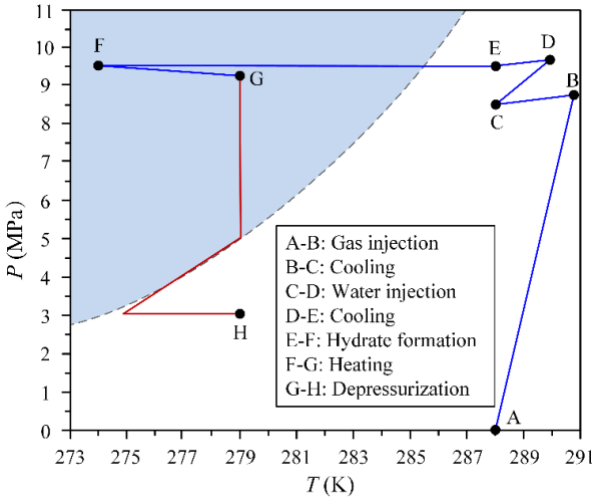

Fig. 2 Trajectory of the pressure and temperature within the reactor during hydrate formation (in blue line) and depressurization (in red line).

2.4 Data processing

During the depressurization process, the hydrate reservoir is eroded by the overlying water, simultaneously the reservoir is producing water. The real-time volume occupied by methane in the reactor during depressurization, $V_{g,rea}$ (t) is presented as:

$$V_{g,rea}(t) = V_{rea} \cdot \varphi - V_{w1} - V_{w2} - v \cdot t + m_w(t)/\rho_w \quad (1)$$

where V_{rea} is the effective volume of the reactor excluding the volume of the wellbore, φ represents the porosity of sediments, V_{w1} represents the volume of water injection (C-D stage of Fig. 2), V_{w2} represents the volume of water injection during hydrate formation (E-F stage of Fig. 2), v is the water injection rate during depressurization, t represents the duration of the depressurization, $m_w(t)$ represents the real-time water production obtained by the precision balance, ρ_w denotes the density of water. The amounts of moles of methane in the reactor, $n_{g,rea}(t)$ can then be calculated as:

$$n_{g,rea}(t) = \frac{V_{g,rea}(t)}{\upsilon(P_1, T_1) M_g} \quad (2)$$

where $\upsilon(P_1, T_1)$ is the specific volume of methane at P_1 and T_1, M_g is the molar mass of methane gas.

The real-time gas volume in gas-water separator and gas collector during depressurization, $V_{g,col}$ (t) can be presented as:

$$V_{g,col}(t) = V_{col} + V_{sep} - m_w(t)/\rho_w \quad (3)$$

where V_{col} is the volume of gas collector, V_{col} is the volume of gas-water separator, and ρ_w denotes the density of water. The amount of gas in the gas-water separator and gas collector, $n_{g,col}$ (t) can then be calculated as follows:

$$n_{g,col}(t) = \frac{V_{g,col}(t)}{\upsilon(P_3, T_2) M_g} \quad (4)$$

where $\upsilon(P_3, T_2)$ is the specific volume of methane at P_3 and T_2.

During gas production from hydrate-bearing sediments with overlying water, the real-time amounts of dissociated gas from hydrates, $n_{g,dis}$ (t) is calculated from Eq.(5):

$$n_{g,dis}(t) = n_{g,rea}(t) + n_{g,col}(t) - n_{g,rea}(t_0) \quad (5)$$

where t_0 indicates the last minute of the hydrate formation stage. According to the chemical formula of MH ($CH_4 \cdot 5.75H_2O$), the amount of dissociated hydrate is equaled to $n_{g,dis}$. With the quantification of the total amounts of dissociated gas from hydrates, the hydrate saturation S_h can then be found as:

$$S_h = \frac{n_{g,dis} M_h}{\rho_h \varphi V_{rea}} \quad (6)$$

where M_h represents the molar mass of MH, ρ_h denotes the density of MH (0.91 g/cm^3).

3. RESULTS AND DISCUSSION

To synthesize MHs in sediments, the water source is continuously pumped to the reactor at a constant pressure to simulate the water supply from the overlying water above the sediments for MHs formation. While the water source is continuously pumped to the reactor at a constant flow rate to simulate the stream from the overlying water above sediments during the entire hydrate dissociation process, thus enabling to investigate the effects of overlying water on the production characteristics of hydrate-bearing sediments.

3.1 Formation characteristics of MHs in sediments with overlying water

The pressure and temperature profiles during formation stage are shown in Fig. 3(a). The flow rate of water injection and hydrate saturation is presented in Fig. 3(b). As shown, the formation process of MHs with overlying water can be divided into three stages. In Stage I, under constant pressure conditions, the reservoir temperature drops below the hydrate phase equilibrium. In Stage II, the signals of the sudden increase in temperature and water injection flow rate indicated that large amounts of MHs are formed. In Stage III, continue to consume gas and water slowly due to hydrate formation. The hydrate saturation and water saturation of the synthesized sediment sample at the end of the formation stage were 44.1% and 53.8%, respectively. Compared with our previous work [6, 7], to obtain samples with similar three-phase saturation, the overlying water shortened the hydrate formation time by more than 3/4.

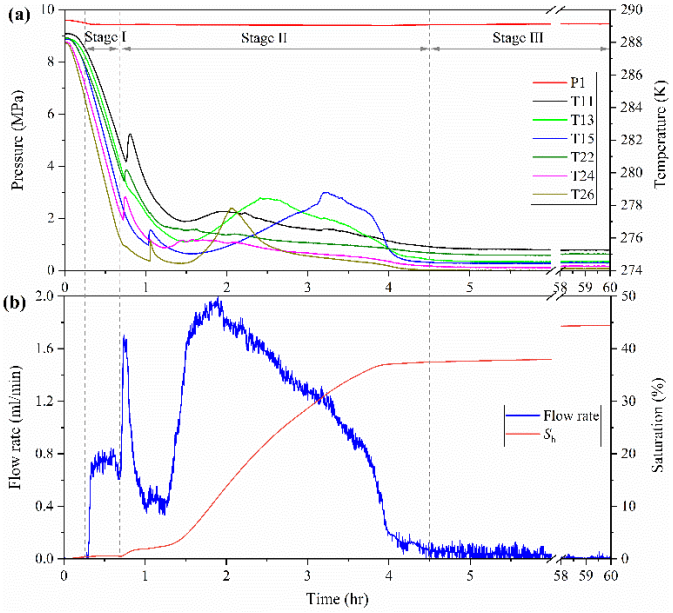

Fig. 3 (a) Pressure and temperature, and (b) flow rate of water injection and hydrate saturation during the entire formation stage.

3.2 Dissociation characteristics of MHs in sediments with overlying water

After the stage of hydrate formation, steady-rate depressurization was conducted to produce gas from MH-bearing sediments with overlying water. Fig. 4(a) shows the typical evolution of pressure and temperature during depressurization. It is noted that the bottom pressure (P_2) of the reservoir begins to drop when the top pressure (P_1) drops to near the hydrate phase equilibrium, and the reservoir pressure difference reaches 4.5 MPa. This is because the high hydrate saturation leads to the low permeability of the reservoir. Depressurization causes the average temperature of the reservoir to approach the hydrate phase equilibrium (see G-H stage of Fig. 2). Because the six-point thermocouple T_2 is close to the overlying water erosion port, when the reservoir pressure is decreased to the exploitation pressure (3 MPa), its average temperature is 0.9 K higher than that of T_1.

The cumulative gas and water production from hydrate-bearing sediments with overlying water during depressurization are presented in Fig. 4(b). As shown, the liberation of free gas leads to the initial pressure drop. Subsequently, the steady-rate depressurization was mainly dominated by water production until lower than hydrate equilibrium pressure. Hydrate dissociation leads to improved reservoir permeability, which allows hydrate dissociation gas and water to be produced rapidly. Compared with our previous work [6, 7], overlying water contributes to hydrate dissociation and gas-water production during depressurization.

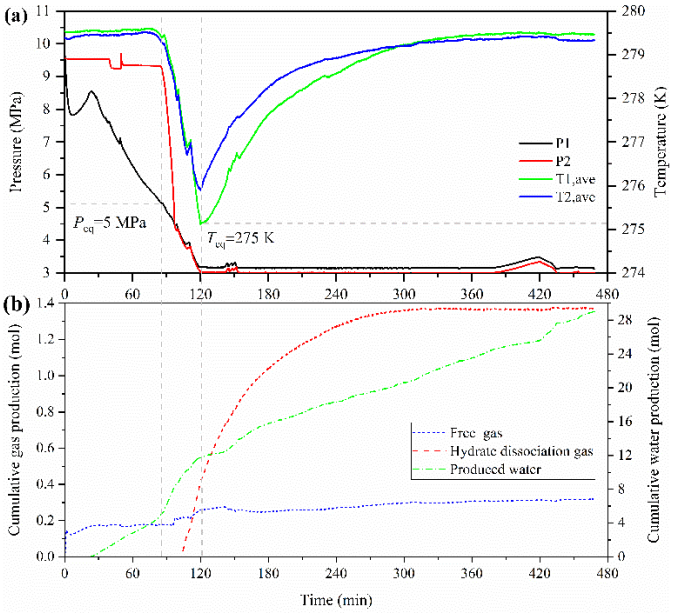

Fig. 4 (a) Pressure and temperature profiles, and (b) gas and water production during depressurization.

4. CONCLUSIONS

In this study, the effects of overlying water on the formation and dissociation of MHs in sediments were analyzed. The results indicate that overlying water can provide a sufficient water source and driving force for hydrate formation. Additionally, overlying water contributes to heat and mass transfer, thereby promoting hydrate dissociation and gas-water production during depressurization.

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