

# Analysis of Gas Recovery and Heat Transfer Properties during Methane Hydrate Dissociation in Frozen Sandy Sediments

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## ABSTRACT

In order to find out the relationship of the heat transfer and the dissociation of permafrost gas hydrate in porous media, this study has conducted a hydrate dissociation experiment below freezing point in a cuboid pressure vessel. The dependences of gas production and hydrate dissociation on the following heat fluxes are also analyzed through numerical simulation: the heat conducted across the boundary  $Q_B$ , the heat injected from the well  $Q_{inj}$ , the sensible heat change of the deposit  $Q_S$ , the heat absorbed by hydrate dissociation  $Q_H$ , the latent heat of ice melting  $Q_L$ , and the unutilized heat  $Q_U$ . The results show that the dissociation process of frozen gas hydrate can be divided into three steps: ice melting, hydrate dissociation, and ice regeneration. The existence of solid ice shows strong inhibition effect on hydrate exploitation.  $Q_H$  mainly comes from  $Q_{inj}$ , and the heat transferred across the boundary is the main component of  $Q_U$ . It implies that the heat injection is the dominating driving force for hydrate dissociation below freezing point.

**Keywords:** gas hydrates, heat transfer, depressurization, thermal stimulation, freezing point

## 1. INTRODUCTION

Natural gas hydrate (NGH) is a kind of solid compounds formed by water and small gas molecules (such as  $CH_4$ ,  $C_2H_6$ , and  $CO_2$ ) under high pressure and low temperature conditions [1]. It is proved that huge amount of gas hydrate exists in submarine and in the permafrost areas, and the total quantity of carbon trapped in NGH is about twice as much as that in

conventional fossil fuels [1]. Thus, it is widely considered to be a potential energy source which can solve the urgent problem of energy shortage of our mankind [2]. Aiming at producing gas from hydrate deposits, various dissociation methods have been investigated, such as depressurization [3], thermal stimulation [4], inhibitor injection [5], and gas exchange method [6].

As the dissociation of gas hydrate is an endothermic reaction which needs to absorb huge amount of heat, the heat transfer is a key factor controlling its dissociation efficiency [7]. Zhao et al. [8] found that the dissociation was affected by the sensible heat, which was related to the porous media material and the water content. Liu et al. [4] concluded that utilizing the heat transferred from the environment was important for reducing the energy consumption. Tupsakhare et al. [9] found that the free water in the pores played an important role in taking the heat to distant hydrate locations. Yin et al. [10] analyzed the influencing factors of spatially heterogeneous methane hydrate dissociation by depressurization. It was determined that the dissociation rate was limited by the heat transfer from the surroundings. Liu et al. [11] numerically analyzed the heat and mass transfer with different overburden and underburden temperatures of hydrate deposits. The results indicated that the impact of thermal conductivity on hydrate dissociation was more pronounced when no external heat was supplied.

Although a general comprehension of the effect of heat transfer on hydrate dissociation has been obtained in these studies, they lack quantitative characterizations of various heat fluxes in hydrate deposits as they are difficult to be measured directly in the experiment. In addition, the presence of ice is rarely considered in the previous studies as it is thought to be able to hinder the

gas production rate by blocking the flow channels of the porous media [12]. However, the evolution of ice is quite common in the permafrost-associated frozen hydrate deposits, such as the gas hydrates discovered in northern Alaska [12] and in Qinghai-Tibet plateau permafrost [13]. It is extremely important to quantitatively analyze the effect of ice on the heat transfer process during gas production from frozen sediments, which can provide insightful knowledge for the optimization of gas production strategies.

The aim of this study is to investigate the heat transfer behaviors during the dissociation of frozen gas hydrate in porous media. Methane hydrate is dissociated below freezing point by depressurization and wellbore heating in a cuboid pressure vessel (CPV). The evolution of various heat flows and their relationships are quantitatively calculated through numerical simulation. In addition, the phase transition properties of ice are also discussed.

## 2. EXPERIMENTAL AND NUMERICAL SIMULATIONS

### 2.1 Experimental apparatus and procedure

The schematic of the cuboid reactor is shown in Fig 1. Its inner effective volume is 1.5 L ( $100 \times 100 \times 150$  mm). The working pressure and temperature of the reactor are 0 to 25 MPa and  $-5$  to  $30$  °C, respectively. The internal temperature of the vessel is monitored by 9 thermometers (T1 – T9). The system pressure is measured by two pressure transducers connected to the inlet and outlet pipelines of the reactor. There is a vertical well at the axis of the vessel for gas production, and its length is 120 mm. Wellbore heating can be realized by connecting an electrical power with the  $73.5 \Omega$  resistance heating wires in the inner surface of the well. Other details of this system can be found in the literature [14-16].

In order to conduct the dissociation experiment, methane hydrate samples are firstly prepared in the reactor, which is filled by quartz sands with particle size of  $0.27 - 0.38$  mm. A total of  $3.86$  mol methane gas and  $361.81$  ml deionized water are injected into the reactor for hydrate formation at an environmental temperature of  $7.80$  °C. When the pressure is decreased to  $11.00$  MPa, the formation process is completed. Then the free gas and water are released from the vessel to reduce the system pressure to  $5.50$  MPa. After that, the environment temperature is adjusted to  $-1.00$  °C. When all the liquid water is transformed into solid ice, the production process is started with two steps: (i) free gas releasing step, in which the pressure is decreased to  $1.50$

MPa; (ii) gas production step; in which the wellbore heating ( $50$  W) is adopted under constant production pressure of  $1.50$  MPa. This study only investigates the second step with wellbore heating. Table 1 shows the experimental parameters and results of hydrate dissociation below freezing point in step ii.

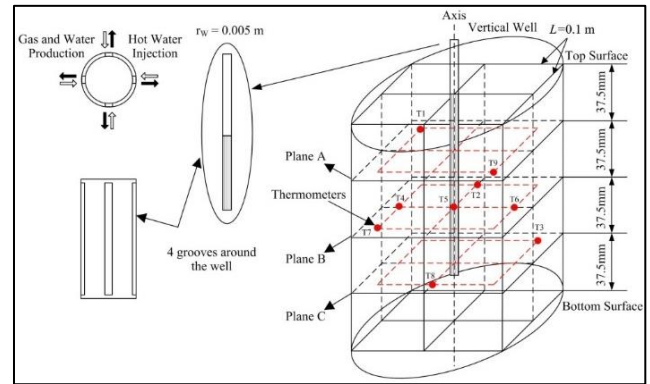


Fig 1 Schematic of the experimental reactor.

Table 1. Experimental conditions and results of hydrate dissociation below freezing point.

Parameter	Value
Volume of the reactor	1.5 L
Gas composition	CH <sub>4</sub>
Porosity of quartz sand	46.0%
Initial phase saturations	$S_{H0} = 0.511$ , $S_{I0} = 0.113$
Wellbore heating rate	50 W
Production pressure	1.50 MPa
Boundary temperature	$-1.00$ °C
Production time	242.20 min
Volume of produced gas	60.156 L

### 2.2 Numerical simulation

The parallel version of TOUGH+HYDRATE (T+H) code is employed and modified for the numerical simulation. It is specifically set up for the description of methane hydrate formation and dissociation characteristics in complex geologic media. Previous studies have proved the effectiveness of this code for modeling the hydration reactions, phase transition, and flow of fluids and heat in hydrate-bearing sediments [2, 16].

The three-dimensional grid described in the study of Wan et al. [16] is used to represent the CPV in the numerical simulation. The porosity of each element is set to be  $0.460$ . The initial saturations of hydrate, gas, and ice are set to be  $S_{H0}$ ,  $S_{G0}$ , and  $S_{I0}$ , respectively, as shown in Table 1. The boundary temperature is maintained constant at  $-1.00$  °C. During the production process, the pressure of the uppermost wellbore grid is fixed at  $1.50$  MPa, and the other wellbore elements act as source terms, of which the total heat injection rate is  $50$  W.

### 3. RESULTS AND DISCUSSION

#### 3.1 Gas production and hydrate dissociation

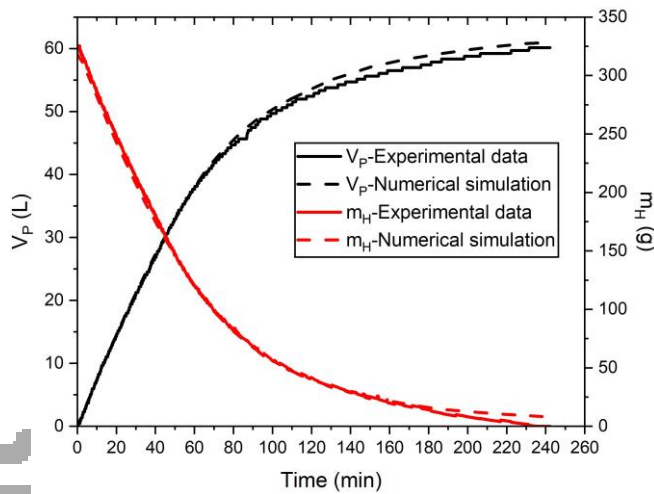


Fig 2 Profile of the total volume of the produced gas  $V_P$  and the remaining mass of gas hydrate  $m_H$ .

Fig 2 shows the experimental and numerical simulation results of the total volume of the produced gas  $V_P$  and the remaining mass of gas hydrate  $m_H$  during hydrate dissociation below freezing point. It can be seen that  $V_P$  increases continuously during the whole simulation period, which indicates the effectiveness of the employed method for the exploitation of frozen gas hydrate. However, the rising rate of  $V_P$  tends to slow down due to the gradually declined hydrate dissociation rate. In addition, the heat absorption efficiency of gas hydrate will also be decreased with the decline of  $m_H$ , as more and more injected heat will be absorbed by the quartz sand and other materials in the vessel. The simulated curves of  $V_P$  and  $m_H$  both agree well with the corresponding experimental data, which implies the reliability of the modified kinetic model for the prediction of gas hydrate decomposition in the reactor. The deviations observed in the later production process may be caused by the uneven distribution of gas hydrate. As the gas hydrate will be more likely to be deposited in the upper area of the vessel [17], the injected heat will be more easily absorbed by the gas hydrate in the experiment due to smaller distance between the dissociation interface and the wellbore.

#### 3.2 Profiles of the remaining ice

Fig 3 shows the numerical results of the evolution of the remaining mass of ice during gas hydrate dissociation below freezing point. In the beginning, a total of 67.16 g ice is distributed uniformly at the surface of the hydrate

particles. In other words, the gas hydrate is completely frozen by solid ice. The presence of these ice will strongly prevent the dissociation of gas hydrate due to its self-preservation effect [18]. Once the wellbore heating is activated, the ice deposited in the vicinity of the well would absorb the injected heat quickly, which leads to the decline of the ice mass. At the same time, once the gas hydrate particles which are previously trapped by the ice are exposed to the low-pressure environment (1.50 MPa), they will be dissociated very fast, and the released water from hydrate dissociation will be transformed into solid ice. In other words, the dissociation process of frozen gas hydrate can be divided into the following steps: ice melting, hydrate dissociation, and ice regeneration. The injected heat plays an important role in breaking the protection effect of ice on hydrate dissociation. As the dissociation front expands far away from the well, the reduction rate of ice tends to decline with time.

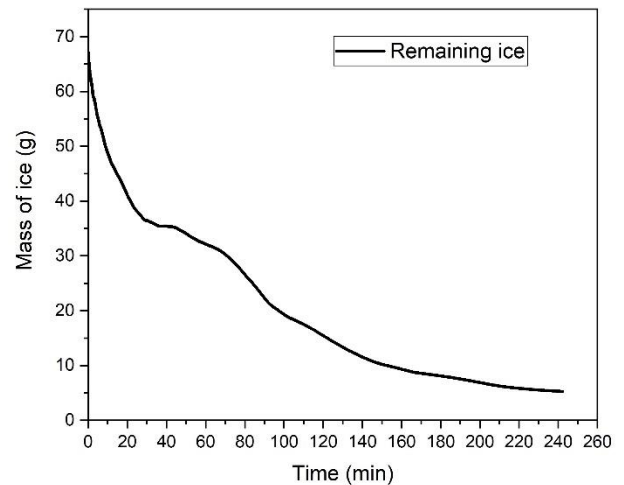


Fig 3 Numerical results of the evolution of the remaining mass of ice in the deposit.

#### 3.3 Analyses of heat transfer behaviors

Fig 4 presents numerical results of the evolution of the cumulative heat conducted across the boundary  $Q_B$ , the heat injected from the well  $Q_{inj}$ , the sensible heat change of the deposit  $Q_S$ , the heat absorbed by hydrate dissociation  $Q_H$ , the latent heat of ice phase transition  $Q_I$ , and the unutilized heat  $Q_U$  during hydrate dissociation below freezing point. The calculation method can be found in the published literature [16]. It is observed that  $Q_B$  decreases persistently in the whole simulation period, which indicates huge amount of heat loss across the low-temperature boundary. On the other hand,  $Q_{inj}$  rises linearly due to the constant wellbore heating rate (50 W). So the injected heat plays a dominating role in promoting the dissociation of frozen gas hydrate. Part of  $Q_{inj}$  will be

absorbed by the porous media and stored as sensible heat. So the  $Q_S$  also increases all the time. The curve of  $Q_H$  shows a continuous rising tendency, which is in accordance with the  $m_H$  discussed in Fig 2.

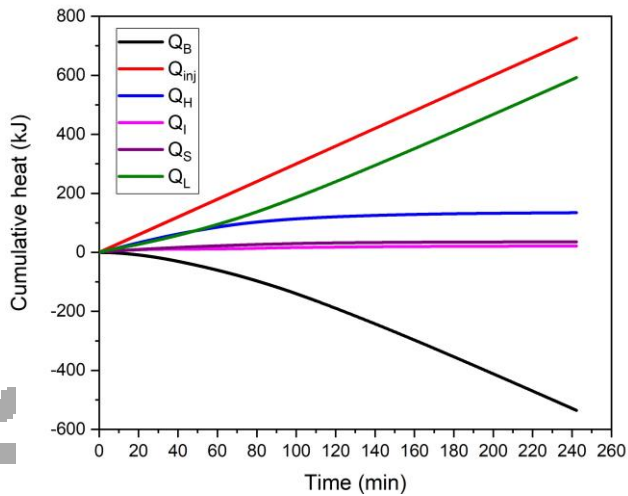


Fig 4 Profiles of the various heat flows during hydrate dissociation below freezing point.

Compared with other heat flows, the curve of  $Q_L$  tends to change more moderately. This is caused by the limited amount of solid ice in the pores. Furthermore,  $Q_L$  is mainly composed of  $Q_B$  and  $Q_S$ . So the curve of  $Q_L$  increases continually to a level even higher than  $Q_H$ . Most of the injected heat is lost across the boundary, and the effect of  $Q_L$  on hydrate dissociation is relatively small when compared with  $Q_{inj}$  and  $Q_B$ .

#### 4. CONCLUSIONS

The gas production and heat transfer behaviors during the dissociation of frozen gas hydrate have been analyzed in this study. The ice deposited in the vicinity of the well would absorb the injected heat quickly. At the same time, the gas hydrate will be dissociated very fast, and the released water from hydrate dissociation will be directly transformed into solid ice. So the dissociation process of frozen gas hydrate can be divided into three steps: ice melting, hydrate dissociation, and ice regeneration.

The injected heat plays a dominating role in promoting the dissociation of frozen gas hydrate. The effect of the latent heat of ice on hydrate dissociation is relatively small. The lost heat is mainly composed of the heat transferred across the boundary as well as absorbed by the porous media.

#### ACKNOWLEDGEMENT

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#### REFERENCE

- [1] Chong ZR, Yang SHB, Babu P, Linga P, Li XS. Review of natural gas hydrates as an energy resource: Prospects and challenges. *Appl Energy* 2016;162:1633-52.
- [2] Li B, Li XS, Li G, Feng JC, Wang Y. Depressurization induced gas production from hydrate deposits with low gas saturation in a pilot-scale hydrate simulator. *Appl Energy* 2014;129:274-86.
- [3] Wang B, Fan Z, Wang PF, Liu Y, Zhao JF, Song YC. Analysis of depressurization mode on gas recovery from methane hydrate deposits and the concomitant ice generation. *Appl Energy*. 2018;227:624-33.
- [4] Liu S, Liang YP, Li B, Wan QC, Han X. Interaction relationship analysis between heat transfer and hydrate decomposition for optimization exploitation. *Fuel* 2019;256.
- [5] Li G, Wu DM, Li XS, Zhang Y, Lv QN, Wang Y. Experimental investigation into the production behavior of methane hydrate under methanol injection in quartz sand. *Energy Fuels* 2017;31:5411-8.
- [6] Sun YF, Wang YF, Zhong JR, Li WZ, Li R, Cao BJ, et al. Gas hydrate exploitation using  $CO_2/H_2$  mixture gas by semi-continuous injection-production mode. *Appl Energy* 2019;240:215-25.
- [7] Zhao JF, Liu D, Yang MJ, Song YC. Analysis of heat transfer effects on gas production from methane hydrate by depressurization. *Int J Heat Mass Tran* 2014;77:529-41.
- [8] Zhao JF, Wang JQ, Liu WG, Song YC. Analysis of heat transfer effects on gas production from methane hydrate by thermal stimulation. *Int J Heat Mass Tran* 2015;87:145-50.
- [9] Tupsakhare SS, Kattakola S, Castaldi MJ. An application of the results from the large-scale thermal stimulation method of methane hydrate dissociation to the field tests. *Ind Eng Chem Res* 2017;56:4588-99.
- [10] Yin ZY, Moridis G, Chong ZR, Tan HK, Linga P. Numerical analysis of experimental studies of methane hydrate dissociation induced by depressurization in a sandy porous medium. *Appl Energy* 2018;230:444-59.
- [11] Liu JJ, Shao ZL, Wu MY, Zheng YX. Heat and mass transfer analysis of depressurization-induced hydrate decomposition with different temperatures of over- and underburden. *J Nat Gas Sci Eng* 2017;44:65-76.
- [12] Moridis GJ, Silpngarmert S, Reagan MT, Collett T, Zhang K. Gas production from a cold, stratigraphically-

bounded gas hydrate deposit at the Mount Elbert Gas Hydrate Stratigraphic Test Well, Alaska North Slope: Implications of uncertainties. *Mar Pet Geol* 2011;28:517-34.

[13] Li XS, Li B, Li G, Yang B. Numerical simulation of gas production potential from permafrost hydrate deposits by huff and puff method in a single horizontal well in Qilian Mountain, Qinghai province. *Energy* 2012;40:59-75.

[14] Li B, Liu SD, Liang YP, Liu H. The use of electrical heating for the enhancement of gas recovery from methane hydrate in porous media. *Appl Energy* 2018;227:694-702.

[15] Liang YP, Liu S, Zhao WT, Li B, Wan QC, Li G. Effects of vertical center well and side well on hydrate exploitation by depressurization and combination method with wellbore heating. *J Nat Gas Sci Eng* 2018;55:154-64.

[16] Wan QC, Si H, Li B, Li G. Heat transfer analysis of methane hydrate dissociation by depressurization and thermal stimulation. *Int J Heat Mass Tran* 2018;127:206-17.

[17] Wan QC, Si H, Li G, Feng JC, Li B. Heterogeneity properties of methane hydrate formation in a pilot-scale hydrate simulator. *Appl Energy* 2020;261:114325.

[18] Chuvilin E, Bukhanov B, Davletshina D, Grebenkin S, Istomin V. Dissociation and self-preservation of gas hydrates in permafrost. *Geosci* 2018;8.