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# Hydrate dissociation induced by depressurization in conjunction with warm brine stimulation in cubic hydrate simulator with silica sand

Jing-Chun Feng [a,b,c,d,1], Yi Wang [a,b,c,1], Xiao-Sen Li [a,b,c,*]

[a] Key Laboratory of Gas Hydrate, Guangzhou Institute of Energy Conversion, Chinese Academy of Sciences, Guangzhou 510640, PR China
[b] Guangdong Key Laboratory of New and Renewable Energy Research and Development, Guangzhou 510640, PR China
[c] Guangzhou Center for Gas Hydrate Research, Chinese Academy of Sciences, Guangzhou 510640, PR China
[d] University of Chinese Academy of Sciences, Beijing 100083, PR China

## HIGHLIGHTS

- Depressurization & warm brine injection is applied for hydrate dissociation.
- Effect of injected salinity on heat transfer, gas production, and energy ratio is analyzed.
- Optimal injected salinity in this work is 10%.

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## ABSTRACT

To study the effect of salt concentration of brine injection on hydrate dissociation, hydrate dissociation experiments induced by depressurization in conjunction with warm brine stimulation have been carried out in a Cubic Hydrate Simulator (CHS). The dual horizontal wells were set as the well configuration. The results indicate that the salinity in the reservoir decreases continuously during the depressurizing stage under the mixture of fresh water from hydrate dissociation. However, the salinity increases overtime during the constant-pressure stage (the injection stage) by the mass transfer with the injected brine. The gas production rate and heat-transfer rate for pure water injection are lower than those for brine injection. In addition, raising the injected salinity can enhance the rates of heat transfer and gas production when the salinity is lower than 10.0%. However, the promotion effect of brine injection on hydrate dissociation is limited when the injected salinity is beyond 10.0%. This is because the specific heat of the brine declines with the increase of the salinity, which causes the decrease of heat injection rate. The water production rate equals to the water injection rate in the process of brine injection. The energy analysis and the evaluation of energy ratio indicate that the optimal injected salinity in this work is 10.0%.

## 1. Introduction

Investigation into new sources of energy is imperative under the conditions of depleting oil resource and the contaminations of burning fossil fuels [1]. For the past few years, natural gas hydrate has been considered to be a strategic and potential energy by both the developing countries and the developed countries [2]. This is due to the extensive distribution of the natural gas hydrate in the permafrost regions and the deep marine areas. In addition, the high energy density of the gas hydrate is the other advantage over the conventional energy resource [3]. Methane gas is the most common gas trapped in the natural gas hydrate reservoir. The previous study indicates that simple $CH_4$-hydrates concentrate methane volumetrically by a factor of 164 when compared to standard pressure and temperature conditions [4]. Moreover, for the past few decades, gas hydrate has been widely used in the technology of cold storage [5], energy storage [6,7], gas-hydrate engine [8], and $CO_2$ separation [9].

Unlike the conventional resources of oil and gas, natural gas hydrate exists as the state of solid in the natural environment of high pressure and low temperature. To recover gas from the natural gas hydrate-bearing sediment, the precondition is the in situ dissociation of hydrate [10]. Over the past few years, the methods of depressurization [11–15], thermal stimulation [16,17], inhibitor stimulation such as the chemical [18,19] or brine injection [20,21],

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[*] Corresponding author at: Key Laboratory of Gas Hydrate, Guangzhou Institute of Energy Conversion, Chinese Academy of Sciences, Guangzhou 510640, PR China.
*E-mail address:* lixs@ms.giec.ac.cn (X.-S. Li).
[1] These authors contributed equally to this work.

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as well as the carbon dioxide replacement [22,23] have been investigated by the researchers through both the numerical and experimental approaches. Each method has its advantage and disadvantage. For example, the depressurization is well known for the merit of no extra energy input, however, the depressurization is unfavorable for the drawbacks that the reservoir is required to have high permeability and the sensible heat of the reservoir is always insufficient for hydrate dissociation. Moreover, blockages of ice formation and hydrate reformation are associated with the single depressurization [24]. Thermal stimulation can effectively solve the problems of ice and reformed hydrate blockage. However, the single thermal injection confronts the disadvantages of severe heat losses in the reservoir and the wellbore [25]. Thus, the previous studies show that the depressurization in conjunction with thermal stimulation is a profitable method for hydrate dissociation and it takes advantage of the synergistic effect of depressurization and thermal stimulation [26–28]. Salt is a thermodynamic inhibitor for hydrate formation, which shifts the phase equilibrium of gas hydrate to higher pressure and lower temperature. The modeling research of Kamath and Godbole [29] showed that the brine stimulation had the advantages of lower energy requirements for reservoir heating and hydrate dissociation, higher gas production rate, reduced heat losses, and improved thermal efficiency over the hot water injection or steam stimulation. It is easy to acquire brine compared to other chemical inhibitors such as methanol and ethylene glycol. Moreover, recently, Chen et al. [30] proposed a novel method for preparation of warm brine in situ seafloor and used the warm brine for the exploitation of gas hydrate. Some literatures reported about the experiments of hydrate dissociation by brine injection. Kamath et al. [31] carried out a range of hydrate dissociation experiments by brine injection and depressurization with a hydrate core holder. They concluded that hydrate dissociation was a function of brine temperature, brine injection rate, brine concentration, and the contacting area of brine and hydrate. Li et al. [32] experimentally investigated the hydrate dissociation by hot brine injection with the middle temperature range (−1 to 130 °C) in a one dimensional reactor. They reported that the hydrate dissociation rate increased with the increase of brine salinity in a certain range, whereas too high brine concentration was helpless for hydrate dissociation. Later on, Lee [33] experimentally studied the hydrate dissociation performance under the stimulation of brine injection in a one dimensional reactor filled with porous rock. They reported that excessively high concentration of brine resulted in the decline of gas production rate remarkably, which was because the excessive NaCl molecules perturbed the fluid flow in the pores and reduced the permeability of the reservoir. In summary, the brine concentration is an important factor, which influences the gas production rate when the hydrate experiments were conducted with brine stimulation. The previous works were mainly focused on one dimensional experimental investigation. Nevertheless, the three dimensional characteristics of heat and mass transfer were not well understood. In addition, the pressure of the previous experiments ranges from 4 to 6 MPa, which is smaller than the pressure of the hydrate reservoir in the South China Sea. Hence, the experiments, which represent the pressure and temperature conditions of the practical reservoir, are urgently required for the guidance of the exploitation of gas hydrate in the South China Sea. In addition, there is lack of the investigation of the multi-well production behavior from the hydrate-bearing reservoir.

In this work, a series of hydrate dissociation experiments under the stimulation of brine injection with different brine concentration were carried out in a cubic three dimensional reactor. For this work, depressurization in conjunction with warm brine stimulation using dual horizontal wells is firstly applied in the experimental research. The conditions of pressure and temperature for hydrate formation and dissociation were based on the geological features of the hydrate-bearing sediments in the South China Sea. The dual horizontal wells were performed as the well configuration, and the temperature of the injected brine was according to our previous study of the optimal temperature of thermal injection for hydrate dissociation in the sandy reservoir [34]. The changing characteristics of pressure and temperature, the behaviors of gas and water production, as well as the characteristics of heat and mass transfer, were obtained. In addition, the characteristic of warm brine (38–39 °C) injection for hydrate dissociation in sandy reservoir was well understood.

## 2. Experimental section

### 2.1. Experimental apparatus

Fig. 1 shows the schematic of the experimental system in this work, which has been introduced in detail in the previous studies [26,35]. The Cubic Hydrate Simulator (CHS), which is the core component of the experimental system, was immersed in a temperature controllable thermostat. The CHS was made of stainless steel 316, and it can withstand the maximum pressure of 30 MPa. As shown in Fig. 1, there's each a gas flow meter at the inlet and the outlet to measure the amount of the injected and produced gas, respectively. Two pressure transducers were placed at the top and bottom of the CHS, respectively. The production pressure was controlled by a back-pressure regulator which was settled at the outlet value. The produced fluid was separated by the gas/liquid separator, and the amount of produced water was weighed by a balance. The injected brine was prepared in advance and it was stored in the middle containers. The heating and injection rate of the brine were controlled by a heater and a metering pump, respectively.

Fig. 2 shows the internal structure of the CHS and the well spacing pattern in this work. As seen in Fig. 2, the inner space of the CHS is uniformly divided into 4 parts by three horizontal layers (Layer A, Layer B, and Layer C). The previous works show that there are a total of 9 vertical wells and 9 horizontal wells in the CHS [28,36]. In this work, the injection well (Well HC) is placed on the middle axle of the Layer C. Well HA, which is situated on the Layer A, is performed as the production well. In this work, grooves are evenly distributed along the whole injection well and production well. The hot water or hot brine injected into the CHS and the gas and liquid produced from the CHS are through these grooves. As shown in Fig. 2, a total of 25 thermocouples evenly distributed on each layer, and the distance between each thermal couple and the adjacent thermal couple is 45 mm. Therefore, there are a total of 75 ($25 \times 3 = 75$) thermocouples in the CHS. The naming of the thermocouples obeys the following rule: as an example, the thermocouples at point 25 in Layer A, B, and C are named as T25 A, T25 B, and T25 C, respectively.

### 2.2. Experimental process

The experimental process mainly includes the hydrate formation and dissociation process. The detailed information of hydrate formation is shown in Table 1. Before hydrate formation, the CHS was cleaned up and dried. Then the CHS was tightly filled up with silica sands with the diameter of 300–450 μm. The intrinsic permeability of the porous media was approximately 50 D [37]. Afterward, the preparative brine solution with the salinity of 2.7% was injected into the CHS. The brine is made using sodium chloride, purity grade 99.5%, from Xilong Chemical Co., Ltd. The salinity of the liquid in the CHS after hydrate formation is shown in Table 1. It shows that the end salinities of the brine for runs 2–6 are