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# Fluid flow mechanisms and heat transfer characteristics of gas recovery from gas-saturated and water-saturated hydrate reservoirs

Yi Wang [a,b,c,d,1], Jing-Chun Feng [e,1], Xiao-Sen Li [a,b,c,d,*], Yu Zhang [a,b,c,d], Zhao-Yang Chen [a,b,c,d]

[a] Guangzhou Institute of Energy Conversion, Chinese Academy of Sciences, Guangzhou 510640, PR China
[b] CAS Key Laboratory of Gas Hydrate, Guangzhou 510640, PR China
[c] Guangdong Provincial Key Laboratory of New and Renewable Energy Research and Development, Guangzhou 510640, PR China
[d] Guangzhou Center for Gas Hydrate Research, Chinese Academy of Sciences, Guangzhou 510640, PR China
[e] School of Engineering, Sun Yat-sen University, Guangzhou 510275, PR China

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ABSTRACT

Due to the huge reserves, natural gas hydrate is considered as a potential energy resource in future. Therefore, developing methods of gas recovery from hydrate reservoirs for commercial production are attracting extensive attention. In this work, hydrate dissociation and gas recovery from the gas-saturated and water-saturated hydrate accumulations are investigated in a pilot-scale hydrate simulator. Depressurization, thermal stimulation, and depressurization assisted thermal stimulation method are adopted in this work. Furthermore, the mechanisms of fluid flow and the heat transfer during hydrate dissociation in different hydrate accumulations are elucidated by large-scale experimental results. The experimental results indicate that the fluid flow mechanisms and the heat transfer characteristics during the gas recovery from hydrate reservoirs are greatly influenced by the initial water saturation. The Optimum gas production method is also different for different hydrate accumulations. The depressurization is optimized method for hydrate dissociation in the gas-saturated reservoir considered from the aspect of gas-water ratio. Thermal stimulation results in the lowest gas-water ratio and the lowest hydrate dissociation ratio, and is not effective for both the gas-saturated and water-saturated hydrate reservoir. The depressurization assisted thermal stimulation is the optimum method for the hydrate dissociation in the water-saturated sample.

## 1. Introduction

Within the framework of sustainable development, energy supply and energy security are important factors for both the developed countries and developing countries. Natural gas hydrate is a potential clean energy resource due to the huge reserves of natural gas trapped in hydrate bearing formations. Although the precise estimation of natural gas hydrate on the earth is uncertain and the estimation vary from $2.8 \times 10^{15}$ to $8.0 \times 10^{18}$ $m^3$ [1,2] the amount of natural gas trapped in gas hydrate is huge. The common perception is that the total carbon content in gas hydrate is more than twice as much as that of all of the conventional fossil fuels [3]. The energy density of methane hydrate is 2–5 times larger than that of the conventional natural gas, and 10 times larger than the other kinds of unconventional gas sources, such as shale gas and coal bed gas [4]. Therefore, developing methods for commercial gas recovery from natural gas hydrate are attracting extensive attention.

Hydrates are crystalline clathrate compounds composed of a lattice of water molecules and encasing one or more guest molecules [5]. In nature, methane is the most common guest molecule for natural gas hydrate. Natural gas hydrate exists under conditions of high pressure and low temperature, which occur in the deep oceanic sediments and the sediments within the permafrost regions [6]. Given the fact that natural gas hydrates occur in solid state and special pressure-temperature conditions, gas production from natural gas hydrate reservoirs requires in-situ hydrate dissociation generally. This means that natural gas hydrate must be converted into its component gas and water in a controlled way [7]. Over the past few decades, depressurization [8,9], thermal stimulation [10,11], inhibitor injection [12], carbon dioxide replacement [13], and the combined application of these methods have been extensively applied and investigated for hydrate dissociation [14]. The models of hydrate decomposition with different methods

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* Corresponding author at: 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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are also reported in our pervious work [15,16]. In order to observe hydrate dissociation in the real environment and assess the feasibility of exploitation technologies for commercial production, a series of field tests on gas production from hydrate reservoirs have been carried out. The field test in the Mallik region of Canada in 2008 proved that gas production from hydrate accumulations was technically feasible from a sand-dominated reservoir, which promotes the potential of hydrates to be considered as an important recoverable energy resource [17]. During a field test in the Prudhoe Bay area (the north slope of Alaska/USA), the methane exchange by $CO_2$ injection into a methane hydrate reservoir has been tested for the first time [18]. Not only hydrate field tests of gas production from hydrate accumulations in the permafrost regions, but also the marine hydrate field tests were successfully conducted in the Nankai Trough by Japan in 2013 [19] and in the Shenhu area of South China Sea by China in 2017 [20]. In both of these marine hydrate field tests, the depressurization method is applied for gas recovery.

Because field-scale hydrate production tests are associated with the features of long-time preparation, huge cost and high risk, limited field tests were carried out up to date. Experimental investigations play a significant role for the study and development of key exploitation technologies and to clarify the corresponding principals. In earlier times, Ohmur et al. [21] used a 0.036 L reactor to study the morphology of hydrate formation and dissociation. Later on, Kono et al. [22] carried out the methane hydrate dissociation experiments in a 0.188 L batch reactor. In addition, a 5.8 L cubic hydrate simulator was employed for three dimension analyses of the hydrate dissociation behaviors [23,24] and the optimization of thermodynamic parameters [25]. Pang et al. used a middle-sized (10 L) reactor to study methane hydrate dissociation using thermal stimulation. They showed that the heat transfer rate and the thermodynamic driving force were the key rate-limiting factors for hydrate dissociation in the closed reactor [26]. Fitzgerald et al. [27] applied a 59.3 L reactor to investigate the methane hydrate dissociation performance by thermal stimulation. Their research showed that higher initial hydrate saturations resulted in better production performance and larger heating rates caused higher peak efficiency rates. Because the reservoir scale influences the fluid flow and heat transfer in the hydrate reservoir, several large reactors have been built in several laboratories recently. For example, Li et al. [28] have built a 117.8 L hydrate simulator. They applied this simulator to investigate the hydrate dissociation and gas production behaviors [29,30], the heat transfer and mass transfer characteristics [30], the influence of well configuration [31], and the entropy production features [32]. Funded by the German national research project SUGAR, hydrate dissociation experiments were executed successfully in a 425 L large reactor [13]. The biggest system to date is the High-pressure Giant Unit for Methane-hydrate Analyses (HiGUMA) in Japan with an inner volume of 1710 L. Hydrate dissociation experiments under one step and multi-step depressurization have been performed in this simulator. The results indicated that an appropriate heat of the hydrate-bearing sediments was a crucial factor for inducing hydrate dissociation [33].

The saturations of hydrate, gas, and water are very important factors influencing the hydrate dissociation process. In nature, hydrates are generally coexisted with free water or free gases. Directly at the base of gas hydrate stability (BSHS) all three phases can coexist. Based on the differences of coexisting phases, reservoirs can generally be subdivided into three classes. Class 1 describes the situation that a hydrate bearing layer (HBL) is overlaying on a two-phase fluid zone with free gas, which is considered to be the most suitable hydrate reservoir for natural gas recovery. Class 2 is characterized by a mobile water zone below the HBL, while Class 3 describes the situation where the HBL lies between layers without mobile fluid phases. Most of the hydrate reservoirs in marine sediments are labeled as Class 3 [34]. Various international research projects are intended to investigate the hydrate reservoirs associated with the highest potential for commercial production [35]. The majority of the laboratorial experiments on hydrate dissociation are carried out under the conditions of high gas saturation (Class 1). However, hydrates in marine environments mainly represent Class 3 [36]. In this work, the gas-saturated hydrate-bearing samples and the water-saturated hydrate-bearing samples are investigated. Moreover, the dissociation of originally gas-saturated or water-saturated hydrate-bearing samples may be different. For instance, Konno et al. [37] found that ice formation rate was faster in a water-saturated hydrate reservoir when conducting the methane hydrate dissociation experiments in sandy porous sediment below the quadruple point. To date, there are limited reports with respect to the systemic analysis of gas production from the gas-saturated and water-saturated hydrate-bearing reservoir, especially with different production methods.

Beyond this background, methane hydrates are synthesized in sediments in a pilot-scale hydrate simulator (PHS). By using the PHS, our previous works have employed to develop the efficient and sustainable gas production methods in the hydrate reservoir. The coupled phase change process, heat/mass transfer, and gas/water production characteristics during hydrate dissociation by different methods can be investigated by the large-scale experiments. Serials of the innovative methods for gas production from hydrate were investigated in these large-scale experiments, for example, the deep depressurization method, the depressurization combined with warm water stimulation method, and the five-spot thermal huff and puff method. Furthermore, the dissociation method conditions can be optimized by the experimental investigations, such as the production pressure, the heat injection temperature, and the heat injection rate. However, the hydrate-bearing samples in these researches were basically gas-saturated samples. In this study, hydrate dissociation and gas recovery from the gas-saturated and water-saturated hydrate accumulations by different production methods are firstly investigated by large-scale experimental system. Depressurization, thermal stimulation, and depressurization assisted thermal stimulation method are adopted in this work. The fluid flow mechanisms and heat transfer characteristics during hydrate dissociation in different hydrate accumulations are elucidated. Furthermore, the optimized methods for the gas recovery from different hydrate accumulations are concluded.

## 2. Experiments

### 2.1. Experimental apparatus

The experimental system mainly consists of 6 primary subsystems: (A) a high-pressure reactor which is made of stainless steel 316 served as the core component; (B) a gas and liquid injection system including the gas pump, the gas flow meter, and the metering pump for water; (C) a production control system which comprises the gas/liquid separator, the back-pressure regulator, the gas flow meter and balance; (D) an ambient temperature controlling system; (E) a data acquisition system that records the pressure, temperature as well as gas and water flow parameters, and (F) measuring units. The schematic of the experimental system is shown in Fig. 1. The experimental system has been described in all details in the previous study [30]. The high-pressure reactor is named the “Pilot-scale Hydrate Simulator” (PHS). The inner of the PHS is cylindrical with the inner diameter of 0.50 m and the inner height of 0.60 m. The effective inner volume of the PHS is