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Influence of gas sweep on methane recovery from hydrate-bearing sediments



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HIGHLIGHTS

- Gas sweep is in favor for reducing the partial pressure of methane in free gas.
- Methane recovery rate by N₂ injection is much quicker than direct depressurization.
- The batch N₂ injection mode is more suitable for controlling hydrate dissociation rate.
- The method of N₂ sweep is quite suitable for low saturation hydrate reservoir.

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ABSTRACT

Gas sweep is favorable for reducing the partial pressure of methane in free gas during the production of methane from hydrate-bearing sediments. To evaluate the influence of gas sweep on methane recovery, pure N₂ was injected into hydrate-bearing sediments using a scale-up three-dimensional apparatus. The influences of injection mode, hydrate saturation, and N₂ injection rate were investigated. The experimental results suggest that the methane recovery rate by pure N₂ sweep is much quicker than direct depressurization. The driving force for hydrate dissociation increases with the increase of the N₂ mole fraction, which further promotes the decomposition of hydrates and guarantees a high gas production rate. In terms of N₂ injection rate on gas production, the higher the gas production, the more N₂ that is required, which means the increase of gas production rate is at the cost of injecting much more N₂. Compared with the continuous mode, the batch injection mode is more suitable for controlling the hydrate dissociation rate and may be the lower risk way for hydrate exploitation. The gas sweep method may supply a new strategy that would be helpful to make low saturation hydrate reservoirs become a technically recoverable resource.

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1. Introduction

Natural gas hydrates are non-stoichiometric crystalline solids formed by water and natural gas under low temperature and high-pressure, which widely exist in subsea sediments and permafrost zones (Sloan and Koh, 2007). The gross reserves of organic carbon bound in natural gas hydrates is conservatively estimated to be double that of all known fossil fuels on the earth (Makogon, 2010; Boswell and Collett, 2011). Therefore, as an unconventional resource of natural gas, gas hydrates are considered as an important energy

source in the near future and have attracted much attention from researchers all over the world. However, it is an enormous challenge to extract natural gas from gas hydrates currently. Some methods for producing natural gas from hydrates have been proposed, such as depressurization, thermal stimulation, chemical inhibitors injection, and CO₂ or mixed gas (CO₂/N₂) replacement. Gas hydrates production via thermal stimulation involves the increase of temperature above the hydrate stability region by technologies such as hot brine injection, steam or cyclic steam injection, which has been investigated experimentally (Tang et al., 2005; Tsimpanogiannis and Lichtner, 2007; Linga et al., 2009; Yang et al., 2010) and numerically (Moridis et al., 2002; Pooladi-Darvish, 2004; Tonnet and Herri, 2009). This method suffers from a vast loss of heat used to improve the temperature of hydrate-bearing geologic reservoir. Gas hydrate

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production via depressurization is considered to be the most economically promising technology for no charge extra energy (Ji et al., 2001; Moridis and Collett, 2003; Moridis and Sloan, 2007; Liang et al., 2010; Yang et al., 2012; Oyama et al., 2012). However, the temperature and driving force for hydrate dissociation drop in response to the depressurization of hydrate-bearing sediments, leading to a gradual decrease in the production rate, which is ultimately controlled by heat transfer toward the hydrate dissociation region (Lee et al., 2010; Seol and Myshakin, 2011; Li et al., 2014a). In addition, if the pressure difference between equilibrium and outlet is high, the pores will become plugged by hydrates regeneration (Nagao, 2012). Therefore, extra energy is still needed to sustain the hydrate dissociation in the depressurized operation. Injecting thermodynamic inhibitors can result in hydrate dissociation as the temperature and pressure conditions for hydrate stability are shifted (Sung et al., 2002). The most common thermodynamic inhibitors are alcohols (e.g., methanol, mono-ethylene glycol, and di-ethylene glycol) and salts (e.g., NaCl, CaCl₂, KCl, and NaBr), however their use in the production of natural gas hydrates is limited by environmental impact and high economic costs. The injection of CO₂ into hydrate-bearing sediments can cause the release of CH₄ and the formation of CO₂ hydrate within the CH₄ hydrate stability field, which serves as a dual purpose for the recovery of an energy source and greenhouse-gas sequestration. The CO₂–CH₄ replacement has been systemically investigated including the feasibility of kinetics and thermodynamics (Zhao et al., 2012; Komatsu et al., 2013), replacement mechanism (Yoon et al., 2004; Bai et al., 2012; Qin and Kuhs, 2013), and influence of CO₂ phase type (gaseous, liquid, or CO₂ emulsion) (Ota et al., 2005, 2007; Yuan et al., 2012, 2013, 2014). However, the CO₂–CH₄ replacement method suffers from a low replacement rate for industrial production (Ota et al., 2005). In order for gas hydrates to become a producible energy resource, new technologies and approaches are still needed.

Compared with gas or liquid CO₂ replacement, methane recovery driven by gas mixture, for example, CH₄–N₂/CO₂ (Park et al., 2006; Koh et al., 2012), CH₄–air or CH₄–CO₂/air (Kang et al., 2014), has been investigated and seem to be more feasible in terms of cost and environmental protection, which had also been adopted by US DOE in 2012 to verify the capacity of CH₄–CO₂ replacement by injecting N₂ (77%)+CO₂ (23%) into hydrate-bearing sediments at Ignik-Sikumi field (NETL, 2012). However, at the initial stage of gas injection, gas mixtures would diffuse in hydrate reservoir firstly, rather than replace with methane immediately. In addition, the direct injection of large amounts of gas mixture may accompany with thermal stimulation for the sensible heat of gas. As water and hydrates coexist in the pore space, CO₂ may react with free water, thus would likely to improve the concentration of N₂ in the stratum. However, up to now, most researches only focus on studying the influence of CH₄–CO₂ replacement on gas production. To more closely examine the gas production from hydrate-bearing sediments by injecting gas mixtures (CO₂, N₂, air, etc.) in real scenarios in the field, further investigations will consider the effects of gas sweep in pore space and the sensible heat induced by gas mixtures, which may cause the dissociation of gas hydrates.

On the other hand, the gas hydrate stability is controlled by conditions such as temperature, pressure, salinity, and gas composition (Li et al., 2014b). The first three parameters have been investigated to evaluate the capacity of gas production except for gas compositions. Recently, Kinnari et al. (2014) supplied some hydrate management strategies in practical gas and oil production systems, including gas sweep. In this work, to examine the capacity of changing gas composition on methane production, a series of experiments using a scale-up three-dimensional apparatus were conducted to investigate the influence of N₂ sweep on methane recovery from hydrate-bearing sediments. Compared with CH₄ and CO₂, N₂ is much more difficult to form hydrates

which can avoid blocking the mass transfer channels. It also has lower solubility in water, and is easily to be extracted out from the seafloor. Being the main compositions of coal-bed gas, methane–nitrogen mixtures can be separated by mature technologies, such as pressure swing adsorption (Fatehi et al., 1995) or adsorption by MOFs or ZIF-8 (Liu and Smit, 2009, 2010). The market price per unit volume of methane is approximately 10 times of nitrogen in China, which makes this method be feasible in terms of economy. Besides, the influence of operation mode (the batch injection mode and the continuous injection mode) and other operation factors, such as the N₂ injection rate, the injection–production ratio, and hydrate saturation, were also investigated in this work.

2. Experimental section

2.1. Materials

Methane and nitrogen with purity of 0.999 were supplied by the Beijing Beifen Gas Industry Corporation, China. The brine (NaCl) solution with salinity of 33.5 g/L was prepared in the laboratory. The hydrate-bearing sediments with a porosity of 0.387 were formed by 20/40 mesh quartz sands with an average diameter of 0.38 mm.

2.2. Apparatus

A schematic diagram of the experimental apparatus is shown in Fig. 1, which has been described in our previous work (Yuan et al., 2012, 2013, 2014). It mainly consists of a high-pressure reactor, a gas injection system, a cooling system, a gas collection system, a gas–water separation system, and a data acquisition system. All experiments are conducted in a high-pressure reactor constructed from stainless steel with an effective volume of 7.05 L ($\phi 300$ mm \times 100 mm) and maximum working pressure of 16 MPa. The reactor is placed into a water bath containing ethylene glycol solution to maintain a constant temperature ranging from 253 to 353 K. Sixteen thermocouples, with an accuracy of 0.1 K, are inserted into the reactor from the top to detect the temperature distribution and variation during hydrate formation and gas sweep process, which are divided into four groups along the radial direction. The distribution of the thermocouples (T1–T16) in the reactor is shown in Fig. 2. Pressures are monitored by two pressure transducers with an accuracy of 0.02 MPa, which are mounted on the top and the bottom of the reactor, respectively. A Monitor and Control Generated System (MCGS) is used to collect and record data of temperature, pressure, and flow rate during the experiment. A filter is attached to the front of mass flow transducer to prevent water from affecting the accuracy of mass flow.

2.3. Procedures

The experiments were conducted according to the following procedures, which are divided into two parts: the preparation of CH₄ hydrate-bearing sediments and the N₂ sweep process.

2.3.1. Preparation of hydrate-bearing sediments

The method of CH₄ hydrate preparation for each group of replacement experiment is the same as that used in our previous study (Yuan et al., 2012). First, a known amount of brine solution was cooled to 273.2 K and quartz sands were frozen to 267.2 K, and kept for 24 h. Then, the brine solution was injected into the sands, and stirred immediately and adequately, so that water can be in the form of fine ice particles homogeneously distributed in the sand. The ice–sand mixture was filled in the reactor before each experiment. To ensure that the water remained as ice in the

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