Applied Energy 187 (2017) 180–188

Contents lists available at ScienceDirect

Applied Energy

journal homepage: www.elsevier.com/locate/apenergy

Gas recovery from depressurized methane hydrate deposits with different water saturations

Mingjun Yang, Zhe Fu, Lanlan Jiang, Yongchen Song *

Key Laboratory of Ocean Energy Utilization and Energy Conservation of Ministry of Education, Dalian University of Technology, Dalian 116024, China

HIGHLIGHTS

- NGHs deposits with excess gas or water were both recoveried by depressurization.
- Water saturation and depressurization range are key factors for NGHs dissociation.
- Larger depressurization range accelerates NGHs dissociation for excess gas deposit.
- The obvious water mobility in excess water deposits hinders methane gas output.

ARTICLE INFO

Article history:
Received 3 July 2016
Received in revised form 31 August 2016
Accepted 8 October 2016

Keywords:
Methane hydrate
Depressurization
Excess water
Excess gas

ABSTRACT

Natural gas hydrates (NGHs) are new and clean energy resources with significant potential. Many studies have investigated NGHs in an attempt to recover natural gas from NGHs deposits. Additional investigations are still needed to clarify the dissociation characteristics of NGHs to develop safe and efficient recovery methods. In this study, two types of NGH deposits were simulated by forming methane hydrates (MHs) in porous media: the first type was formed with excess gas, and the other type was formed with excess water. The formed MHs were dissociated by depressurization methods. Magnetic resonance imaging (MRI) was used to monitor the liquid water distribution and quantify the MH amounts during formation and dissociation. The results showed that a larger depressurization range enhanced the average rate of MH dissociation and gas production for excess gas conditions. For excess water conditions, the mobility of liquid water was dominant during MH dissociation and hindered methane gas output. Furthermore, a larger depressurization range accelerated MH dissociation. When MH dissociations were compared for various gas-water saturated porous media, liquid water saturation and depressurization range were identified as two key factors affecting MH dissociation.

1. Introduction

Natural gas hydrates (NGHs) are typically found in permafrost and marine sediment, which contains abundant hydrocarbon gas and water at high pressures and low temperatures [1,2]. NGHs have recently been considered as potential non-conventional fossil fuel resources due to their huge reserves [3–5]. To utilize NGHs as an energy source, NGHs are dissociated via phase change processes (i.e., from solid to gas and liquid) and require gas recovery methods. However, to date, no economically viable gas recovery method has been developed for NGH deposits. Due to a number of mechanisms and new technologies still being developed, the exploitation of NGHs has remained at laboratory and short-term production scales. Methane hydrates (MHs) are widespread worldwide and are typically measured in NGH dissociation investigations [6–10]. These studies have primarily focused on developing guidance and recommendations for the safe and efficiency recovery of on-site MH deposits.

The three traditional MH exploitation methods are depressurization, thermal stimulation, and inhibitor injection [11]. Compared with the other two methods, depressurization is considered a more competitive method due to its low energy consumption and low environmental impact [12,13]. For the depressurization process, the pressure of a MH deposit is decreased to levels below the hydrate phase equilibrium pressure to initiate dissociation [14,15]. Many laboratory-scale studies have explored CH_4 recovery methods in MH deposit depressurization [6,16–19]. These investigations have focused on gas production rates, efficiency, and output [7,20]. Other studies have focused on heat and mass transfer characteristics [21]. The gas production characteristics have been reported to be influenced by the compositions and sizes of

* Corresponding author.
E-mail address: songyc@dlut.edu.cn (Y. Song).

http://dx.doi.org/10.1016/j.apenergy.2016.10.029
0306-2619/

reservoirs [16,17]. These experiments have typically used glass beads, silica gels, and artificial sandstone cores to simulate the deposits. Numerical simulations have also been used to model MH recovery by depressurization [22,23,9,24]. Recently, combinations of depressurization with other methods have been proposed for CH_4 recovery [25,26].

Overall, gas recovery from MH deposits is complicated and involves thermodynamic considerations, two phase flows, heat and mass transfer, physics and chemistry [8,27–29]. Hydrate saturation levels and permeability in MH deposits also affect CH_4 recovery [30,31]. Permeability is especially a sensitive parameter [32,33]. Different classes of NGHs deposits are known, and three could be potentially exploited for gas recovery [34–36]. These three types of deposits cover under burden and are covered by over burden. The first class contains a two-phase zone under a hydrate-bearing layer [37]. An underlying water zone is present in the second class [38]. The third class comprises only an isolated hydrate layer [34]. Moridis et al. investigated gas recovery from Class 1 and Class 2 deposits using numerical modeling. They found that gas can be recovered at high rates for long periods of time from Class 1 deposits by depressurization method. Furthermore, the group found that the permeability of the over burden significantly affected gas recovery from Class 2 deposits [35,36,39]. Konno et al. also investigated depressurization-induced gas recovery methods using simulations on oceanic NGHs sediments [40]. It is necessary to verify the simulation results using experimental data. While the experimental investigation are absent for the comparison of gas recovery characters from the three classes NGHs deposits. Meanwhile, the characters of NGHs dissociation from different class deposits are essential for the assessment of NGHs exploitation technology adaptability. That is to say, it is desiderated to investigate the gas recovery characters from different classes NGHs deposits.

As briefly discussed, the depressurization method for MH recovery has been widely studied, and it is surely the potential way. While most of these investigations are single for excess gas deposits or excess water deposits, and it is hard to make comparison between these investigations. And a few reports have compared the use of excess gas or excess water in recovery methods in MH deposits. It is necessary to measure MH dissociation characters from different class deposits under different backpressures, and it is important for the development of depressurization NGHs exploitation technology. The present study simulated NGHs deposits with different phase saturation, and the MH dissociation characteristics for deposits with different initial water and gas saturation levels are investigated. These reported experimental results may provide guidance for the exploitation of different NGH classes.

2. Experiments

2.1. Apparatus and materials

A high-pressure, nonmagnetic, polyimide-based cylindrical vessel (⌀38 × 314 mm, 12 MPa) was used to form MHs. A vessel jacket filled with a circulating Fluorinert solution (FC-40, 3 M Company, USA) was used to regulate the vessel temperature. A thermostat bath (FL300, JULABO technology, Germany) was used to cool or heat the Fluorinert solution. A separate container in the bath was used to maintain the temperature of water injected into the vessel. An MRI system (operating at 400 MHz, Varian, Inc., USA) was used to visualize and quantify the ^1H in liquid water and thereby determine water distribution in the porous media in the vessel. As liquid water reacted to form solid hydrates, changes in the water distribution and therefore, MH formation and dissociation, were detected by the MRI system. A spin echo pulse sequence was applied at TR = 500 ms, TE = 1.63 ms, and slice thickness = 4 mm. The field of view (FOV) was 30 × 30 mm, and the MRI image matrix was 128 × 128 pixels.

Two high-pressure pumps (260D, Teledyne Isco, Inc., USA) were used to inject water and methane gas into the vessel. A backpressure control valve (BP-2080-M, JASCO, Japan) was connected to the vessel outlet to control the backpressure for MH dissociation. The data acquisition system included temperature and pressure transducers, an A/D module (Advantech Co., Ltd., China) and a computer. The vessel pressure was measured to an accuracy of 0.1 MPa using a pressure transducer (Nagano Co., Ltd., Japan). A thermocouple was used to monitor the jacket temperature. BZ-02 glass beads (0.177–0.250 mm, As-One Co., Ltd., Japan) of approximately 35.4% porosity were packed into the vessel to simulate the porous media. Methane gas (99.99%; Dalian Special Gas Co., Ltd., China) was used to form the MHs. Deionized water was used in all experiments. Additional details regarding the experimental apparatus are provided in a previous paper [41].

2.2. Procedure

The glass beads (BZ-02) were tightly packed into the vessel. After a leakage test, the vessel was placed into the magnetic scanning body of the MRI system. Free gas was evacuated from the vessel, and deionized water was slowly injected to saturate the glass beads until a vessel pressure of 6.0 MPa. Once the pressure reached a steady state, the glass beads were considered fully saturated, and an MRI image was obtained to show the MI value for this state. Then, high-pressure CH_4 (approximately 2.5 MPa) was injected to displace some of the pore water to make space for CH_4 to form the hydrates. The amount of displaced water was recorded for each cycle. Then, the thermostated bath was set and kept at 274.15 K, i.e., the experimental temperature for MH formation and dissociation. After the vessel temperature reached a steady state, CH_4 gas was slowly injected until 6.0 MPa. Images were continuously obtained using MRI.

After the MH formation process, two experimental procedures were tested. The first procedure simulated a natural MH deposit using excess gas. The vessel was depressurized to initiate MH dissociation. The second procedure simulated natural MH deposits using excess liquid water to discharge some of the residual gas before MH dissociation. During the injection process, the water was maintained at 273.65 K to avoid hydrate dissociation. After approximately 5 min, water injection (at 5 mL·min^{-1}) was stopped when excess water flowed out of the vessel and when the vessel pressure was steady. The amount of injected water was approximately twice the pore volume but did not completely displace the residual gas [42]. Based on MH phase equilibrium data (i.e., approximately 2.98 MPa at 274.45 K) [43] and on our previous work [44], the vessel backpressures were set to 2.8, 2.6, 2.4, and 2.2 MPa to dissociate MH deposits. Details of the experimental parameters are shown in Table 1.

3. Results and discussions

Two experimental processes were investigated in this study and are defined by the different water saturation levels in the MH deposits. Case 1 utilized excess gas conditions with approximately 15–17% water saturation. Case 2 utilized excess water conditions with approximately 42–50% water saturation. Four different backpressures were explored for MH dissociation. The MRI system monitored an arbitrary sagittal plane in the vessel to determine the liquid water distribution during MH formation and dissociation in both processes. MH saturation, which is defined as the pore vol-