Chemical Engineering Science 168 (2017) 344–351

Contents lists available at ScienceDirect

Chemical Engineering Science

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

Methane hydrate reformation in porous media with methane migration

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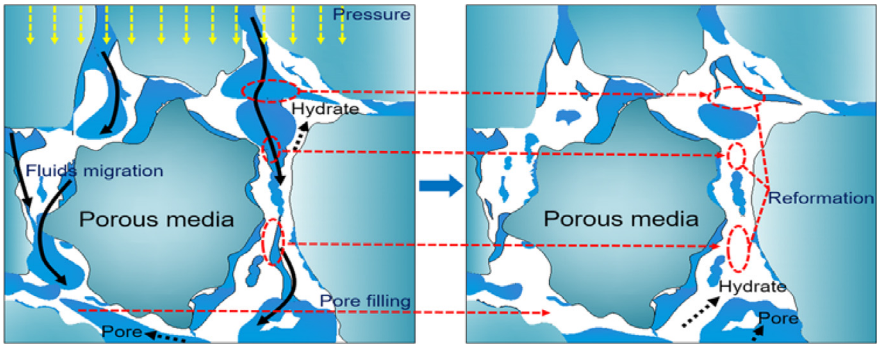
HIGHLIGHTS

- Reformation S_h increment and MH dissociation percentage are positively correlated.
- Percentage of reformation S_h increment increases with methane injection rate.
- S_{w2} is the main factor influencing the hydrate reformation amount and R_{max}.
- Constant methane flow in MH reformation process influence water distribution.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 15 December 2016
Received in revised form 15 March 2017
Accepted 23 April 2017
Available online 25 April 2017

Keywords:
Gas hydrate
Exploration
Reformation
MRI

ABSTRACT

The hydrate reformation that occurs in natural gas hydrate (NGH) exploration reduces mining efficiency and safety. To elucidate the hydrate formation/reformation characteristics during NGH exploration, methane hydrate (MH) was formed/reformed in two different modes to simulate mining of NGH sediment. The effects of residual water, residual MH and methane flow rate on MH reformation in a porous medium were investigated experimentally. Magnetic resonance imaging (MRI) was used to analyze MH saturation and distribution in the porous medium. In reformation, a positive correlation exists between the hydrate saturation (S_h) increment and MH dissociation. Moreover, the percentage of reformation S_h increment increases with the methane injection rate. That demonstrates MH dissociation by depressurization improves the contact area of gas-liquid and enhance the nucleation rate, which contributes to hydrate reformation. In addition, the residual S_w and MH reformation rate maximum (R_{max}) are positively correlated in the rapid-reformation period. According to MRI images, crack-like pathways exist in the porous medium after MH dissociates completely in the first experimental mode. However, constantly flowing methane in the MH reformation process can render the water distribution uniform after MH dissociation in the second experimental mode. That means the methane flow affects the capillary force distribution then further influences the pore water distribution in porous medium.

1. Introduction

Gas hydrates are ice-like crystals in which water molecules (host molecule) form a lattice via hydrogen bonding, and gas molecules (e.g., methane) fill the spaces between the lattice (Englezos, 1993; Sloan, 1998). With the deterioration of the natural environment, research on clean energy sources, such as natural gas hydrate (NGH), nuclear, wind and solar energy, has been increasing (Goel, 2006; Zhao et al., 2012; Zhao et al., 2009). NGH reserves primarily exist in nature on the continental margins and in permafrost. The carbon content of worldwide NGH resources is estimated to be twice that of the existing coal, oil and natural gas resources in total (Hovland et al., 1997). Hence, technologies

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http://dx.doi.org/10.1016/j.ces.2017.04.036
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for NGH production and hydrate dissociation characteristics in sediments have been investigated by many countries due to the significant resource potential (Daraboina, 2015; Ebi et al., 2014; Kumar Saw et al., 2015).

Three popular extraction methods for hydrate exploitation have been studied: thermal stimulation, depressurization and inhibitor injection (Sloan and Koh, 2007; Sloan, 2003). Due to the high energy consumption of thermal stimulation and environmental issues of inhibitor injection, depressurization is considered the most promising for gas hydrate exploitation in the short term and has been tested at the field scale in the Mallik site of the Mackenzie Delta, Canada (Li et al., 2014; Yamamoto and Dallimore, 2008). NGH production efficiency is commonly influenced by heat transfer, phase change, and multiphase flow, among other factors (Veluswamy et al., 2014; Yu et al., 2014). Hydrate reformation could be caused by free-flowing gas in sediments under local temperature. Additionally, hydrate reformation can reduce the permeability of sediments and result in clogging of the penetration path, and hence, long-term hydrate recovery efficiency could be affected (Haligva et al., 2010; Vafaei et al., 2012; Zhang and Lu, 2014). This study aims to examine MH reformation in two experimental modes such that the results can clarify the effect of various factors on MH reformation using depressurization method. That will offer essential data for gas hydrate reformation control during exploration. Gas production properties and heat transfer under static experiments during hydrate dissociation or reformation have been clarified by certain researchers. However, the key factors affecting natural gas hydrate reformation under dynamic conditions must be further investigated to predict and prevent hydrate reformation during NGH production (Hao et al., 2008; Kang et al., 2009; Lv et al., 2012). In addition, there is a dearth of adequate field data, and thus, laboratory scale experiments are a valuable means of studying hydrate reformation. Therefore, it is necessary to elucidate the hydrate reformation characteristics under dynamic conditions to exploit gas hydrate resources safely and efficiently (Bishnoi and Natarajan, 1996; Talaghat, 2009).

Zhao et al. studied natural gas production properties using the depressurization method in porous media. These researchers observed that hydrate reformation occurs in the interior of porous media near the production well, which means that the heat transferred from the ambient environment and the sensible heat of the reservoir are not sufficiently abundant for hydrate dissociation (Zhao et al., 2015). Seol et al. conducted laboratory experiments with X-ray CT to investigate hydrate reformation. The results indicated that heterogeneous pore networks could influence various hydraulic properties of the hydrate-bearing sediment (Seol and Kneafsey, 2009). Visual experimental studies were also performed by Silin et al., and single- and two-phase flow properties in various pore space geometry tight sands were studied using X-ray CT (Silin et al., 2011). Furthermore, due to the superiority of magnetic resonance imaging (MRI), it is commonly used to study hydrate formation or dissociation in porous media (Baldwin, 2003; Birkedal et al., 2015; Hiraia et al., 2000; Yang et al., 2011). Moreover, due to the limitations of field information acquisition, numerical simulation is necessary to research gas hydrate production (Seol and Myshakin, 2011). Using numerical simulation, hydrate reformation is predicted to occur near wellbores after the original gas hydrate dissociation. The temperature decrease caused by hydrate dissociation and the Joule-Thompson effect can cause gas hydrate reformation (Moridis et al., 2011). Because hydrate reformation has adverse impacts on gas hydrate exploitation, it is essential to verify potential MH reformation under certain conditions that approximate the reservoir scale (Anderson et al., 2011; Reagan et al., 2010).

The purpose of this study was to examine MH reformation in two different experimental modes such that the results can clarify the effect of various factors on reformation S_h and rate under dynamic conditions. This information could be used to predict and prevent MH reformation. This research offers safety guidelines for gas hydrate exploration and aids in improving the efficacy of hydrate exploration. MH formation and reformation processes were measured with MRI, and MRI was used to image the ^1H contained in liquid water (Gao et al., 2005; Haber et al., 2015; Yang et al., 2013). Given that the signal sensitivity of ^1H can't be detected in hydrate crystals, MRI can be used to effectively distinguish between liquid phase and solid gas hydrates (Baldwin et al., 2009; Cha et al., 2015). The effects of methane flow rate, residual MH saturation and residual water saturation on the MH reformation process were investigated, and vessel pressure, hydrate distribution, reformation S_h and reformation rate were analyzed in this study. This analysis was performed to obtain critical data that can aid in controlling hydrate reformation during gas hydrate exploration and also improve hydrate exploration efficiency.

2. Experimental method

2.1. Apparatus and materials

Fig. 1 shows a schematic diagram of the experimental system. A high-pressure vessel made of a nonmagnetic material (polyimide) was set in the MRI system and was designed to withstand a pressure of 15 MPa. The effective size is 15 (diameter) × 200 mm. The vessel was surrounded by a jacket that cooled the vessel via a circulated coolant. Three high-precision syringe pumps (260D, Teledyne Isco Inc., Lincoln, NE, America) were used to inject methane and deionized water and to control the back pressure in the experiment. The pressure transducer (3510CF, Emerson Electric Co., Ltd., St. Louis, USA) was connected to the vessel, and the pressure signals acquired from the pressure transducer were collected by the A/D module (Advantech Co., Ltd. Milpitas, USA). Thermostat baths (FL300 and FL 25, JULABO, Seelbach, Germany) were used to control the temperature of the high-pressure vessel and the methane injection pump. An MRI system (Varian, Inc., Palo Alto, CA, USA) for visualizing the MH distribution constitutes the core component of the experimental system. The intensity of the magnetic field was 9.4 T, and the resonance frequency of this system was 400 MHz. A standard spin echo pulse sequence was chosen to obtain the 2D proton density weighted images. The sequence parameters are described as follows: echo time (TE) = 13.82 ms, repetition time (TR) = 500 s, image data matrix = 128 × 128, and field of view (FOV) = 40 mm × 40 mm (4.0 mm in thickness). The sequence acquisition time was 1 min for one image. Methane (99.9%) was sourced from Dalian Specialty Gas Co., Ltd., China. The porous medium used in this experiment consisted of glass beads (porosity 36.8%) from as-One Co., Ltd., Japan. Deionized water was used in all experiments.

2.2. Experimental procedures

The experiments consist of two MH formation and dissociation processes, which are used to simulate the MH reformation process during methane exploitation. The first MH formation (shown as "MH formation" in the following) and dissociation process was performed as follow. The high-pressure vessel was packed with glass beads and placed in the center of the magnetic body. The vessel was connected to the experimental system and evacuated using a vacuum pump. Deionized water was injected into the vessel and pressurized to 8.0 MPa to saturate the porous medium. Methane gas was injected to partially displace the solution through the outlet valve. The amount of displaced water was controlled and measured to determine the initial water saturation. The temperature of