



## Energy efficiency analysis of hydrate dissociation by thermal stimulation



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### ARTICLE INFO

#### Article history:

Received 26 November 2015

Received in revised form

15 January 2016

Accepted 13 February 2016

Available online 17 February 2016

#### Keywords:

Methane hydrate

Thermal stimulation

Dissociation

Energy efficiency

### ABSTRACT

The energy efficiency (EE) is a key parameter to evaluate the performance of methane hydrate (MH) dissociation by thermal stimulation. Experiments of MH formation and dissociation have been performed in a 2D reactor to analyze the EE of hot brine injection, and different influencing factors of EE including geological parameters (MH saturation, intrinsic permeability and initial temperature of the reservoir) as well as thermal stimulation parameters (brine temperature, brine injection rate, brine concentration and the amount of injected heat) have been studied. It is shown that the EE grows with the increasing initial temperature of MH reservoir ( $-1\sim 5\text{ }^{\circ}\text{C}$ ), intrinsic permeability ( $100\sim 1200 \times 10^{-3} \mu\text{m}^2$ ) and brine concentration (2%–20%), and the corresponding maximum EE is 5.7, 5.3 and 8.4, respectively. While the EE reaches a peak and then declines when the total amount of injected heat increases from 100 kJ to 1240 kJ (480 kJ for the maximum EE of 6.4), the temperature of injected brine from 30 °C to 50 °C (40 °C for the maximum EE of 5.2), the brine injection rate from 10 cm<sup>3</sup>/min to 25 cm<sup>3</sup>/min (20 cm<sup>3</sup>/min for the maximum EE of 5.1), and the MH saturation from 16% to 64% (48% for the maximum EE of 7.2).

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### 1. Introduction

The MH is a crystalline, non-stoichiometric compound of methane and water molecules, formed under high pressure and low temperature conditions (Sloan and Koh, 2007). MH is widely distributed around the world with large resources, and is regarded as high-quality alternative energy of the 21st century (Pooladi-Darvish, 2004; Lee and Holder, 2001; Moridis, 2002). Therefore, MH exploitation has attracted increasing interest recent years.

Until now, only one commercial exploitation of MH has been carried out, which is in Messoyakha of Russia (Grover et al., 2008; Makogon et al., 2005), by means of depressurization and inhibitor injection. Besides, several MH field tests have been conducted in Mallik of Canada (Fuji and Takayama, 2008; Kurihara et al., 2010), the eastern Nankai Trough of Japan (Yamamoto, 2014), Alaska of the USA (Schoderbek et al., 2012), as well as Qilian Mountain of China (Zhu and Zhang, 2014), with methods including thermal

stimulation, depressurization, inhibitor injection and CO<sub>2</sub> exchange. Since 1980s, numerous theoretical analysis and experimental studies have been carried out in countries like US, Canada, Japan, India and China etc. Bayles et al. (1989) established an analytical model of MH dissociation by cyclic steam injection, which included only the effect of heat transfer while neglected the effect of fluid flow and drew the conclusion that EE was 4.0–9.6. Kamath and Godbole (1987) compared the methods of hot brine injection and steam injection with a mathematical model, and concluded that hot brine injection could reduce thermal loss and therefore improve the EE. Selim and Sloan (1990) presented an analytical model to describe MH dissociation with thermal stimulation in porous media, in which the process of MH dissociation was regarded as a problem of moving dissociation boundary, and got the EE of 6.2–11.4 with the permeability ranging from 0.1 to 0.5 μm<sup>2</sup>. Kamath et al. (1991) carried out MH dissociation experiments by means of hot brine injection, and concluded that brine concentration would promote the dissociation of MH. Sung et al. (2004) performed a MH dissociation experiment by electric heating and concluded that the EE with constant heat injection method was larger than that under preheating method condition. Tang et al. (2006) experimented on MH dissociation by hot brine injection in a

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1D reactor, with the porosity of 30%, injection temperature ranging 100–190 °C, injection rate ranging 2.5–11 ml/min, and concluded that the EE of MH dissociation was 0.38–2.59. Li et al. (2008) investigated the MH dissociation by microwave heating experimentally, and concluded that when the initial temperature was 2 °C, pressure was 4.25 MPa and the heating power was 60 W, the EE was 3.7, which was larger than that by water bath heating. Dong et al. (2008) conducted experiments of propane hydrate dissociation by surface heat exchanger with the surface temperature of 30.9 °C, and got the EE of 2.25–5.58. Cranganu (2009) proposed a conceptual model involving a horizontal well through which the fuel was injected and dissociated gas was produced, and got quite high EE of 60–89. Li et al. (2012, 2014a,b) investigated MH dissociation in a 1D sand packed tube by hot brine injection, with the brine concentration of 2%, permeability of 1.2  $\mu\text{m}^2$ , injection temperature of 60 °C, injection rate of 12 ml/min, MH saturation range of 12%–44%, and got the EE of 3.5–7.4. Wang et al. (2014) established an experimental model with which thermal stimulation with a five-spot well, heat injection, thermal huff and puff combined with depressurization were utilized, and the EE of 3.67–11.23 was got when the porosity was 48%, MH saturation of 31%, injection temperature of 130 °C, injection rate of 40 ml/min.

It could be concluded from the above analysis that theoretical models analysis and laboratory experimental studies are the main methods to calculate the EE, and compared with theoretical models, experimental studied are closer to the actual production conditions, because the former ones are always based on some assumptions which deviates from the reality greatly. However, the EE of experimental results varies a lot, due to the different methods adopted, such as wellbore heating, hot water injection, in-situ combustion, thermal huff and puff, etc., and different experimental conditions like porosity, MH saturation, heat injection temperature and rate, etc. Until now, no systematical research on the influencing factors on the EE of hot brine injection has been done. In this study, a base case was designed to analyze the process of MH formation and dissociation, as well as the EE of hot brine injection, and other 7 cases were used to study the influencing factors, aiming to conduct a systematical study on the EE of hot brine injection, which would provide some theoretical support for feasible analysis of actual MH exploitation by hot brine injection.

## 2. Experimental apparatus and procedures

### 2.1. Experimental apparatus

The experimental system is shown in Fig. 1, which contains 7 modules: 2D high pressure reactor module, gas supplying module, water supplying module, environment simulating module, back pressure regulating module, data collecting module and parameter measuring module.

The 2D high pressure reactor is the main part of the experimental system with working pressure up to 15 MPa, cuboid inside with 350 mm in length and width and 20–60 mm in height, the cover of which is removable, and the volume of the reactor is changeable (the height used in this study is 60 mm). The sand-packing process was as follows: first, a certain sized sand (180  $\mu\text{m}$ –230  $\mu\text{m}$  for the base case) was packed in the reactor layer by layer, until the needed height was achieved; then the cover of the reactor was pressed into the reactor to pre-compact of the sand; next water was pumped into the four hydraulic cylinders which were installed on the four side edges of the reactor (as shown in Fig. 2) used for compacting the sand.

As is shown in Fig. 2 and 16 groups of temperature and electrode probes are symmetrically distributed in the reactor (16 temperature probes in the upper face and 16 electrode probes in the lower

face) to measure the variation of temperature and electrical resistivity in the process of MH formation and dissociation, and 2 pressure probes are distributed in the upper face of the reactor to measure the pressure variation. The precision of pressure sensors is  $\pm 0.025$  MPa, and that of temperature sensors is  $\pm 0.1$  °C, and for resistivity sensors it is  $\pm 0.1 \Omega \cdot \text{m}$ . The precision of the thermostat is  $\pm 0.5$  °C, and that for the back pressure regulator is  $\pm 0.1$  MPa, while for the gas flow meter the precision is  $\pm 1.0\%$  F.S.

### 2.2. Experimental materials and procedures

The base case is taken as an example to show the experimental procedures and conditions. The water used was brine (NaCl solution) with a concentration of 2.0% and CH<sub>4</sub> with purity of 99.9%. The 2D reactor was packed with quartz sand with grain size of 180–230  $\mu\text{m}$ . Then the reactor was saturated with water and the porosity was measured to be 26.9%, and the intrinsic permeability of water was  $380 \times 10^{-3} \mu\text{m}^2$ , which was measured by Darcy's law.

The procedures of MH formation and dissociation are as follows:

- (1) After the reactor was saturated with brine, CH<sub>4</sub> was injected to displace the brine in the reactor until the brine stops flowing out. Meanwhile the volume of gas injected and water driven out was recorded. Then the outlet valve was shut off and CH<sub>4</sub> was kept on being injected to the preset pressure at 8–9 MPa. Next, the inlet valve was shut off and the reactor was cooled down to 1 °C. The MH formation process was completed when the pressure in the reactor no longer declined, which was around 2.9–3.1 MPa.

The MH formation process was repeated for 3–4 times (Lee et al., 2011), to ensure that the MH was uniformly formed in the reactor.

- (2) Then the back-pressure was regulated equal to the system pressure, which is about 2.9–3.1 MPa. Then the inlet and outlet valve were opened and the hot brine with the temperature of 30 °C was injected from inlet to stimulate MH dissociation, at the rate of 15  $\text{cm}^3/\text{min}$ . When no more gas or water flowed out of the reactor, the back-pressure was gradually reduced to the atmospheric pressure to release the residual gas in the reactor. The variation of temperature, pressure, as well as water and gas production rate was recorded every 10 s during the whole MH dissociation process.

## 3. Analysis of MH formation and dissociation

### 3.1. Calculation of MH saturation

The MH saturation is derived by means of volume balance (Li et al., 2009). The initial temperature and pressure before MH formation are  $T_1$  and  $P_1$  respectively, and after the MH formation, the temperature and pressure of the system become  $T_2$  and  $P_2$ . The pore volume of the reactor is treated as constant, thereby the total volume of the gas and water before the MH formation should be equal to that of water, gas and MH after the MH formation:

$$V = V_{w1} + V_{g1} = V_{w2} + V_{g2} + V_{h2} \quad (1)$$

where  $V$  denotes the total pore volume,  $\text{cm}^3$ ;  $V_{w1}$  and  $V_{g1}$  denote initial volume of water and gas respectively,  $\text{cm}^3$ ;  $V_{w2}$ ,  $V_{g2}$  and  $V_{h2}$  denote the volume of water, gas and MH after MH formation,  $\text{cm}^3$ .

It is further assumed that water and MH are incompressible (Feng et al., 2015), and one unit volume of MH could generate 164

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