Experimental study of gas production from hydrate dissociation with continuous injection mode using a three-dimensional quiescent reactor

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HIGHLIGHTS

► Hydrate exploitation is simulated with continuous injection mode using a 3-D reactor.
► An optimal water temperature and injection rate exists for gas production of hydrate.
► Compared with Na_2SO_4 and EG, injecting NaCl solution has the highest gas production.
► The continuous injection mode is more in favor of gas production of hydrate.

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ABSTRACT

The exploitation of methane hydrate formed under the same conditions is simulated experimentally to investigate the gas production behavior at different injected solutions (hot water, saline solution, and ethylene glycol) with continuous injection mode using a three-dimensional quiescent reactor. The influence of hot water temperature, injection rate, and injected solution type on the gas production and the energy efficiency are examined. The results show that the gas production increases with the increased hot water temperature, but the influence of temperature is weakened when it is beyond 313.0 K. An optimal injection rate also exists for the gas production. Hydrate dissociation with NaCl solution has the highest gas production compared with Na_2SO_4 and ethylene glycol when under the same conditions. The comparison of energy efficiency between the continuous injection mode and the interval injection mode shows that the former mode with double wells is more in favor of hydrate dissociation and gas production.

1. Introduction

Natural gas hydrates are crystalline solids composed of water and gas. Typical natural gas molecules include methane, ethane, propane, and carbon dioxide [1]. Extensive deposits of naturally occurring gas hydrates have been found in many regions of the world. Because of the large size of this resource, gas hydrates are considered as a future, unconventional resource of natural gas [2,3]. A variety of simulation methods for producing gas from hydrate have been proposed, such as depressurization, thermal stimulation, chemical injection, and CO_2 replacement. The merits of CO_2 replacement make it as a potential approach to produce natural gas from hydrate, but also a technique that may allow CO_2

sequestration [4,5]. The main drawback of CO_2 replacement is the low replacement reaction rate and replacement ratio [6,7]. Depressurization is considered to be the most effective method for hydrate dissociation. However, if the pressure difference between equilibrium and outlet is high, the pores will be plugged by hydrate regeneration [8,9]. In comparison, the thermal stimulation and chemical injection method are more easily controlled [8], among which the interval injection mode or the continuous injection mode are in general adopted.

The interval injection mode is widely used in the oil industry to enhance oil recovery. Using this mode, hydrate decomposition by hot water or inhibitor injection has been investigated [10–14]. For the continuous mode, hydrate begins to dissociate after the injection of steam/hot water or chemical inhibitor from one-side of the apparatus, and gas and water are continuously released from the other side. With this operating mode, Tang et al. [15]

investigated the hydrate production behavior under thermal stimulation in a one-dimensional setup of 38 mm in diameter and 500 mm in length. Fan et al. [16] studied natural gas hydrate dissociation by presence of ethylene glycol (EG) using a reactor of 3.5 L and found that the dissociation rate depends on the concentration and the flow rate of EG. Using a one-dimensional vessel (30 mm in diameter and 534 mm in length), Li et al. [17] found that the hydrate dissociation rate decreases with the injection of brine and the hydrate dissociation duration is shortened with the increase of the salinity. However, while NaCl concentration is beyond a certain value, the rate has no longer continued increasing. Using the same apparatus of Fan et al. [16], Dong et al. [18] found that the acceleration effects of EG on the dissociation behaviors of propane hydrate are better than that of methanol with the same injecting flux and mass concentration because of the higher density of EG. Lee et al. [19] studied the dissociation behavior of gas hydrate using the steam stimulation and pointed out that the phase of heated fluid should be adequately selected and monitored during injection period according to the reservoir condition. For hydrate exploitation simulation in three-dimensional device, Li et al. [14] investigated the gas production behavior of methane hydrate using the huff and puff method in a 5.8 L cubic pressure vessel. Li et al. [20] developed a 117.8 L pilot-scale hydrate simulator, but only the gas production from methane hydrate by depressurization was investigated. Su et al. [21] built a 196 L large scale three-dimensional simulation device, and hydrate accumulation distribution in gas seeping system was examined up to now.

It could be found from these pre-existing experimental investigations that the hydrate dissociation by thermal stimulation and chemical injection with the continuous injection mode is in general performed in one-dimensional, small reactor. The temperature distribution and flow behavior from these kinds of devices can not been simulated. The obtained results and conclusions may not be in accord with the real three-dimensional behavior of natural gas hydrate dissociation, since the scale-up effect will have to be considered when the experimental results are applied to gas production in natural gas hydrate and the exploitation of natural gas hydrate resources [22]. In this work, gas hydrate dissociation behaviors in porous sediment during brine, hot water, and EG injection with continuous injection mode are conducted in a three-dimensional reactor. The spatial temperature distribution and the flow characteristics of injected solution are examined and the effects of temperature, injection rate, and different injecting solutions on the gas production are investigated. The comparison of the interval and the continuous injection modes on hydrate dissociation is also performed.

2. Experimental

2.1. Apparatus and materials

The experimental set-up used for formation and dissociation of hydrates in this work is similar as that used by Yang et al. [12,23] and Yuan et al. [11,24]. A diagram of the experimental apparatus and arrangements is shown in Fig. 1, which is divided into four parts: reaction system, injection system, gas recovery system, and data collection system. The primary component of the reaction system is the hydrate formation/dissociation reactor, which is 150 mm in internal radius and 100 mm in height with a volume of about 7 L. The maximum operating pressure of the reactor is 16 MPa. There are sixteen resistance thermometers placed at different positions to measure the temperature distribution in the reactor. The detailed arrangements of the thermometers and the injection well in the reactor are shown in Fig. 1(b). The injection system includes a reciprocating metering pump, an electronic balance, and a water heater with thermostatic control. The gas recovery system consists of two gas–water separators, a filter, a back-pressure regulator, and two gas mass flow transducers. The location of the gas production well is on the top of the reactor. The pressure, temperature, and flow rate data are collected by the corresponding transducers in real time during the hydrate dissociation.

Methane used is supplied by the Beijing Beifen Gas Industry Corporation with a purity of 99.99%. Ethylene glycol is supplied by Beijing Modern Eastern Fine chemical with a purity of 99.99%. Sodium sulfate and sodium chloride are supplied by Beijing Beihua Fine Chemical Limited Company, whose purities are both higher than 99.0%. Brine with a salinity of 3.35 wt.% is prepared in laboratory with sodium sulfate. Sodium chloride solution and sodium sulfate solution with 16 wt.% concentration are also prepared in our laboratory with deionized water. Quartz sand of 20 – 40 mesh with an average diameter of 0.38 mm is cleaned and dried before each experiment.

2.2. Procedure and data processing

2.2.1. Hydrate formation procedure

In order to simulate the dissociation of methane hydrate, the first important step is to form representative hydrate-bearing sand sample in each experiment. The procedure for the preparation of sample is the same as that described in our previous work [11,23]. The key point is that the sand is frozen to 267.2 K firstly, and then the brine at 273.2 K is mixed with the frozen sand immediately and adequately. The water-bath temperature is stabilized at 272.7 K to prevent water from migrating during the gas injection. After methane gas is charged into the reactor to the desired pressure, hydrate will form among the sediments. And when the pressure in the reactor keeps constant for 24 h, it is assumed that the hydrate formation is complete.

2.2.2. Hydrate dissociation procedure

After the representative hydrate sample was prepared, the following procedures are used to study the dissociation characteristics of methane hydrate with hot water/inhibitor injection. When the hydrate sample pressure is stable, valves 5, 10, 11 (or 12), and back-pressure regulator 16 are opened and other valves are closed. The pressure in the reactor decreases to be close to the equilibrium value slowly with the back-pressure regulator to make sure that the hydrate in the reactor does not dissociate. After that, valves 6 and 9 in Fig. 1 are opened and the hot water or inhibitor with specific temperature is injected into the reactor at a constant rate with the metering pump. The hydrate will dissociate continuously under the effect of injected solution, and the pressure will increase because of the dissociation of hydrate. When the pressure exceeds the back-pressure, gas will flow out of the reactor from valve 5, and the gas production period starts. After the mass of injected solution reaches the specified value, the metering pump is closed and valves 6 and 9 are also closed. Valve 5 is still kept opened because the hydrate dissociation will still continue after the end of the injection process. When the flow rate is less than 0.05 L/min, valve 5 is closed and the gas production is assumed to be end. The total volume of gas collected after the beginning of the solution injection while the well pressure (P_{well}) is maintained is named as the final cumulative gas production. Afterwards, the back-pressure regulator is set to 0 MPa, and the temperature of water bath is increased to the room temperature to dissociate the residual gas hydrate in the reactor quickly, and then valve 5 is opened and the residual gas is discharged from the reactor. During the whole experimental time, the value of temperature, pressure, and the gas flow rate are all recorded by the computer at 1 s interval.