



Full Length Article

Experimental study on characteristics of methane hydrate formation and dissociation in porous medium with different particle sizes using depressurization



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ABSTRACT

Natural gas hydrate (NGH) is characterized as a potential energy resource. The commercial application of hydrate energy requires further development due to the complexity of hydrate exploitation. The particle size significantly affects the exchange ability of gas and water in the pores of the sediment during hydrate dissociation. In addition, sediment deformation during hydrate decomposition is generally correlated with marine disasters and requires further characterization. In this work, different quartz sand particle sizes were applied to simulate the sediment of hydrate reservoir. According to the data analysis, the formation and gas production rates of methane hydrate decreased with sediment particle size due to the difference of the specific surface area. In addition, the sediments exhibited different deformations after the hydrate dissociation. Smaller sediment particle sizes generated more obvious deformations following methane hydrate decomposition.

1. Introduction

Natural gas hydrate (NGH) is a solid crystalline compound that consists of gas molecules and H₂O molecules. NGH is formed at high pressure and low temperature conditions and are composed of gas molecules, such as CH₄, C₂H₆, SO₂, and CO₂, which occupy the lattice structure formed by H₂O as guests. NGH is natural and predominantly composed of methane, given that its total proportion in NGH reaches around 90%. As a result, NGH is also called methane hydrate due to its ice-like appearance and flammable characteristics, thereby earning NGH the name “flammable ice” [1]. According to the statistics, 1.97 × 10¹⁴ m³ of stored NGH has been detected worldwide, with the total amount possibly reaching 2.0 × 10¹⁶ m³, given that NGH is mainly distributed on permafrost areas and deep sea sediments. NGH has a large energy density. For instance, the decomposition of 1 m³ of methane hydrate can produce 164 m³ of pure methane gas under atmospheric temperature and pressure conditions [2]. In addition, certain qualities such as large available quantities, worldwide distribution, great energy density have deemed methane hydrate as the most important substitute energy source for coal, petroleum, and natural gas in the 21st century. Methane hydrate is abundantly stored in China,

especially in the Pearl River Mouth Basin, the Qilian Mountain tundra, and the Shenhu area, South China Sea [3].

Four types of methods that differ from traditional oil extraction applications are generally executed to exploit methane hydrate, specifically the thermal stimulation, depressurization, chemical inhibitors injection, and CO₂ replacement methods. These methods all respectively release CH₄ by breaking its pressure-temperature balance [4,5], but naturally have their own advantages and disadvantages. Hydrate energy is barely employed in commercial applications given the complexity of its produced NGH exploitation problems and first requires the execution of laboratory-scale simulations. In general, 99% of the total NGH is stored in deep sea sediment. As a result, the simulation of NGH decomposition in deep sea sediments has become the most widely researched topic in this field [6].

Certain sediment properties, specifically permeability, directly influences the efficiency of NGH exploitation, thereby changing the exchanging ability of gas, water, and heat within a system. In addition, NGH exploitation generally results in NGH sediment deformation. Many researchers have examined the mechanical process of NGH decomposition. Zhu et al. [7] used quartz sand, white corundum, and emery to execute deep sea sediment simulations, wherein methane

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hydrate was formed in a reactor with fixed volume and subsequently decomposed by the depressurization method. The results indicated a positive correlation between the hydrate production rate and the thermal conductivity of sediments. Li et al. [8] designed hydrate decomposition experiments with different hydrate saturations, the results of which exhibited efficiency following the execution of the thermal stimulation method at acceptable hydrate saturation levels. Tang et al. [9] employed TOUGH-Fx/Hydrate software to numerically simulate the hydrate decomposition by the depressurization method. The result indicated the suitability of methane hydrate decomposition kinetics for laboratory-scale experiments. In addition, the exchange of water and methane gas was deemed a key factor in deep sea sediments or the landscape layer. Tsimplanogiannis et al. [10] considered the differences of the permeability, particle size distribution, and filling rate of the sediments, and concluded that the permeability of the sediments initiated the decomposition of methane hydrate. Moreover, Kono et al. [11] acquired a hydrate decomposition kinetics equation and reaction order from conducted hydrate decomposition experiments with different sediment particle sizes. The produced equation exhibited a positive correlation between the particle size of the sediments and the production rate. However, no further details were established.

During hydrate decomposition, the exchange of gas and water in sediment also has a significant impact on sediment deformation, which is an extremely important safety measure during hydrate exploitation. Hydrate exploitation from deep sea sediments or the permafrost zone without proper safety measures can result in geological disasters and other secondary dangerous situations. Zhang et al. [12] simulated methane hydrate storage in a centrifugal machine and decomposed it with the thermal stimulation method. The result indicates the presence of transverse and portrait shifting on the sediments. Hyodo et al. [4,13] used a three-axis mechanical instrument to measure the shear strength of hydrate sediments. Kwon et al. [14] designed a hydrate decomposing experiment that combined a centrifugal machine with high-pressure equipment to analyze hydrate deformation before and after decomposition.

This work aims to characterize the differences of various hydrate decomposition characteristics following changes in the particle sizes of sediments.

2. Experiment

2.1. Experimental apparatus

The flow chart of the experimental apparatus is presented in Fig. 1. The reaction kettle is composed of 316 stainless steel, wherein the reaction space has a cubic-like shape and has a total volume of 726 mL. This kettle was sealed with a rubber O-ring setting on the top. As presented in Fig. 2, 27 Pt100 thermocouples were evenly distributed inside the kettle on three layers, specifically the top (A), middle (B), and bottom (C), to achieve better results. Each layer was placed on 9 thermocouples that were measured at a temperature range of 223.15–473.15 K and a currency of ± 0.1 K. The top cover of the vessel exhibited nine wellheads corresponding to the nine measuring points on each layer within the vessel. The present study chose wellhead 5 as chosen as the production well. The other wellheads were sealed.

A thermostatic water bath was placed outside the kettle to ensure stable environmental temperature conditions. A desander was employed throughout the process to intercept particle outlet flow at a bearing pressure range of 0–40 MPa. A back pressure valve (TESCOM, Emerson Electric Co., USA) was employed to manually maintain the pressure at a range of 0–30 MPa at a currency of ± 0.2 MPa. The outlet flow reached the gas-liquid separator and was separated to gas and water, wherein the liquid flowed down to the container through an electronic scale (Santorius Co.) with the concrete vision Santorius BS 2202S (0–2200 g, ± 0.01 g) and the gas flowed to the flowmeter (Seven Star Co.) with vision D07-11CM (0–10 L/min, $\pm 2\%$).

2.2. Materials and methods

The methane gas (99.9% purity) was produced by the Guangzhou Gas Group Co. Ltd., Conghua branch. Quartz sand made by Bandao Silica Sands Co. was used to simulate the sediments. Different material particle sizes were employed, as presented in Fig. 3. The particle sizes of different sands are 240–350 μm , 125–178 μm , and 37–45 μm . The initial porosities of these samples are 49.71%, 51.01%, and 50.67%.

In this work, three runs of depressurization experiments were carried out in sandy sediment. The 729 mL reactor was all fulfilled with the sediments. The temperature of the water bath was set to 281.15 K. Throughout the experiment, the experimental system was kept in a constant-volume condition and no water or solid was produced. The key condition parameters of the experiments are listed in Table 1. The experimental conditions (temperature/pressure) are similar to the geological conditions at the hydrate reservoir in the South China Sea [1].

To fulfill the reactor, silica sand was firstly packed in the reactor. Then the sediments of the two runs were immersed by deionized water, respectively. After setting the sediments, the reactor was sealed by closing the top cover. Then the residual air was driven out of the reactor twice by the injection and release of methane gas. The entire system was placed in the water bath. After that, methane gas was pumped into the reactor to 20 MPa. Then a gradual pressure drop was observed during the hydrate formation. When the pressure dropped to approximately 13.50 MPa, the decomposition experiments were carried out by depressurization. At the beginning of the experiments, predetermined production pressure was set at 4.70 MPa by the back-pressure regulator. Then, the outlet valve was opened to make the pressure decrease to the production pressure. Subsequently, with the production of gas, the hydrate gradually decomposed. The end of the production process was marked by little gas production. After that, the residual gas was entirely released till the pressure inside the reactor recovered to atmospheric pressure. Finally, the reactor was opened by unlocking the clamps around the top cover to observe the sediment deformation. The repeatability of scientific experiments has been reported in our previous work, the error of this experimental result is lower than 5% [16].

Furthermore, because of the uniform specimen, the isothermal boundary, and the smaller ratio of scale and permeability, the difference between the experimental conditions and the reality cannot be ignored. However, by using the experiments, the heat and mass transfer and hydrate dissociation characteristics can be investigated. The effect of particle sizes on hydrate dissociation and gas production in a same hydrate reservoir element can be investigated by using this experimental system.

The saturation of the hydrate was calculated as follows:

$$S_G + S_W + S_H = 1$$

$$S_G = \frac{v_m \cdot n_{m,G}}{V_{pore}}$$

$$S_W = \frac{m_{W0} - N_H (n_{m0} - n_{m,G} - n_{m,W}) M_W}{\rho_W V_{pore}}$$

$$S_H = \frac{(n_{m0} - n_{m,G} - n_{m,W}) M_H}{\rho_H V_{pore}}$$

where S_G , S_W , and S_H represent the saturation of gas, water, and hydrate; v_m represents the specific volume of methane; $n_{m,G}$, $n_{m,W}$, and n_{m0} represent the mole quantity of the total injected methane gas, free methane gas, and methane dissolved in water, respectively; V_{pore} stands for the total pore volume of the sediments assuming they are incompressible and are considered constant; N_H is the hydration number of methane hydrate; M_W and M_H are molar mass of water and methane hydrate, respectively; and ρ_W and ρ_H are the densities of water and hydrate, respectively.

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