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Visualization study on the promotion of natural gas hydrate production by water flow erosion

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ABSTRACT

Natural gas hydrates (NGHs) are a new, clean, and effective energy source with great potential for exploitation. The efficient exploitation of NGHs has been a focus of research worldwide. Water migration in hydrate sediments is an important parameter influencing NGH exploitation. However, there is still little research in terms of visualization studies on the variation of hydrate distribution during the water flow process in hydrate-bearing sediment. Such variation of hydrate distribution and the influence of water migration on methane hydrate (MH) dissociation with different backpressures and water flow rates were systematically and visually analyzed in this study, where the influence of temperature and pressure variation on MH dissociation was completely eliminated. The results showed that the chemical potential difference between the hydrate phase and the aqueous phase caused MH dissociation during the water flow process and that the rate of MH dissociation increased with decreasing backpressure and increasing water flow rate. When the rate of MH dissociation is low, there will be a longer time for the flow channel to appear, vary, and disappear. Based on this conclusion, a new method of water flow erosion to improve NGH exploitation is proposed in this study.

1. Introduction

With the development of human society, the global energy demand is predicted to increase rapidly in the coming decades [1,2]. Nowadays, traditional fossil fuels such as oil and coal comprise approximately 85.9% of the global energy supply [3]. It is essential to find a sustainable alternate energy source to meet the continuously increasing energy demand with the decrease of traditional fossil fuels [2,4]. Natural gas hydrates, which are widely distributed in continental permafrost or marine sediment, are regarded as an alternative energy resource in consideration of their high energy density and purity [3,5,6]. In past decades, natural gas hydrates (NGHs) have attracted worldwide attention for investigation.

NGHs are crystalline solids composed of methane molecules and water molecules under low-temperature and high-pressure conditions [7–9]. The hydrate thermodynamic equilibrium is affected by temperature, pressure, chemical potential differences, and chemical potential correction caused by the dissolution of guest molecules in pure water [10]. All known hydration decomposition methods are based on shifting the thermodynamic equilibrium of the system [11]. There are mainly four methods of NGH exploitation, as follows: (1) depressurization [12–14], (2) thermal stimulation [15], (3) CO_2 replacement [16–18], and (4) inhibitor injection [19–21]. These four methods are mainly based on the temperature and pressure variations.

In past years, some measurements of NGH exploitation have been performed. The depressurization method has been widely used to liberate natural gas from methane hydrate (MH) reservoirs as a cost-effective solution because it does not require external energy [22]. However, the depressurization method has a low methane production rate, and the endothermic hydrate dissociation reaction may result in secondary hydrate formation [23]. In order to study changes during the hydrate dissociation process induced by depressurization, Yousif et al. [24,25] studied the process of hydrate dissociation in Berea sandstone by depressurization, and they derived a three-phase boundary model to describe the dissociation process. Kono et al. [26] studied the kinetic dissociation rate of hydrates using the depressurization method and found that sediment properties had a significant effect on the dissociation rate. Sloan [27] and Konno et al. [28] studied the characteristics of gas production from oceanic MH reservoirs by the depressurization method and both found that the effective permeability and initial temperature of the reservoirs were important factors of gas production. Regarding the other three NGH exploitation methods, Kawamure [29] studied the hydrate dissociation kinetics by dissociating pellet-shaped samples, which mimic naturally occurring hydrates in ocean sediments, with a viscous fluid or pure water at different temperatures. Tang et al. [30] concluded that the gas production rate

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increases with increasing time until it reaches a maximum and then it starts to decrease but that the water production rate remains nearly constant during the gas production process. They also found that the energy ratio of thermal stimulation generation can be affected by the hydrate content of the sediment, the flow rate, and the water injection temperature. Sun et al. [31] suggested that liquids flowing in the well may lose the most heat, resulting in an obvious decrease in the heat impact. Komai et al. [32] measured the replacement rate between CO_2 and CH_4 occurring in a CH_4 hydrate using in situ Raman spectroscopy, and they found that CO_2 hydrates coexisted with the CH_4 hydrate phase and were generated at both the surface and inside the CH_4 hydrate samples and ice crystals. Geng et al. [33] simulated methane dynamics by using carbon dioxide molecules to replace methane molecules in the hydrate in order to assess the possibility of methane reformation. Mutalik et al. [34] carried out a series of experiment on the depressurization and injection of a hot saline solution. Li et al. [35] studied gas production from MHs through injecting hot brine at different temperatures and concentrations, and found that the concentration and temperature changed the gas production yield.

Irrespective of the type of NGH exploitation method, such methods have always been accompanied by water migration. Many investigations have researched the NGH exploitation process, but thus far, there has been no visualization study on the variation of hydrate distribution and the influence of water migration on MH dissociation in hydrate-bearing sediment during the water flow process. In addition, there are few reports systematically analyzing the influence of the chemical potential difference on MH dissociation. Furthermore, it was found that higher water production has a negative influence on the gas production efficiency irrespective of the exploitation method used [36–38]. Reducing water production is hence very important for hydrate exploitation. In this study, in order to avoid high water production, which leads to low hydrate exploitation rate and efficiency, we considered this problem in terms of excess water, chemical potential difference, and the transfer of heat and mass. We proposed a new method of water flow erosion in which the temperature is lower than phase equilibrium temperature and the backpressure higher than phase equilibrium pressure, to completely eliminate the influence of temperature and pressure variations on MH dissociation. The four main purposes of water flow erosion are as follows: (1) inducing and increasing the chemical potential difference between the water and hydrate phases; (2) enhancing the heat and mass transfer of the hydrate reservoir; (3) increasing the salinity of the hydrate reservoir; and (4) inhibiting the secondary hydrate formation. All these purposes are achievable and promote efficient hydrate decomposition. This investigation thus achieved visualization research by MRI, which is considered as a reliable technology to aid hydrate research and has been used for many years [39]. The experiment results may provide guidance for investigating NGH exploitation.

2. Experiments

2.1. Apparatus and materials

A schematic diagram of the experimental system is shown in Fig. 1. The experimental apparatus shown in Fig. 1 was designed to investigate methane hydrate (MH) dissociation during the water flow process. Xiao-Sen Li et al. [40] found that hydrates will dissociate during water injection and that low-speed flow slightly affects the hydrate dissociation. A magnetic resonance imaging (MRI) system (Varian, Inc., Palo Alto, CA, USA) for visualizing the MH dissociation during the water flow process formed the core component of the experimental system. The resonance frequency of this system with a 9.4 T magnet field intensity was 400 MHz. Two-dimensional (2D) proton density-weighted images were obtained by a standard spin echo pulse sequence that was chosen in this study. The sequence parameters were as follows: echo time (TE) = 4.39 ms, image data matrix = 128 × 128, and the field of view (FOV) = 30 mm × 30 mm (2.0 mm in thickness). The sequence acquisition time was 2.14 min for one image.

A high-pressure vessel capable of withstanding a pressure of 15 MPa and made of a nonmagnetic material (polyimide) was used in this study. The effective size of the vessel was 15 mm in diameter and 200 mm high. The high-pressure vessel was surrounded by a jacket, which was used to cool the vessel via a circulator bath. Three thermostat baths (FL300 and FL 25, JULABO, Seelbach, Germany) were used to control the temperature of the MRI machine, the high-pressure vessel, and the injection pump. Three high-precision syringe pumps (260D, Teledyne Isco Inc., Lincoln, NE, USA) were used to inject methane gas and deionized water, and to control the backpressure in the experiment. Two pressure transducers (3510CF, Emerson Electric Co., Ltd., St. Louis, USA) were connected to the input and output of the high-pressure vessel. A differential pressure sensor (Nagano Keiki, Japan, with an accuracy of ± 0.01 kPa) installed at both ends of the reactor was used to acquire pressure difference signals. The signals acquired from the pressure transducer and the pressure difference signals were collected and processed by the A/D module (Advantech Co., Ltd., Milpitas, CA, USA). Glass beads (As-One Co., Ltd., Japan) with particle diameters of 0.177–0.250 mm (BZ02) mm and a porosity of 35.4% were used as the homogeneous skeleton structure of the porous medium. CH_4 (Dalian Special Gases Co., Ltd., China, with a purity of 99%) was used as the gas source. Deionized water was used in all the experiments.

2.2. Hydrate formation

The glass beads were used to tightly fill the vessel. Then the vessel was placed into the center of the MRI magnet and connected to the experimental system. The volume of the vessel was 12.51 mL, and the porosity of the glass beads was 35.4%. Next, deionized water was injected into the vessel under a constant pressure of 1000 kPa after the vessel was vacuumed. Then, the deionized water was pressurized to 6000 kPa to saturate the porous media and this pressure was maintained for 1 h. The deionized water was displaced by high-pressure CH_4 and the volume of displaced water was used to calculate the initial water saturation. Then, the vessel was pressurized to 6000 kPa with CH_4 gas and kept stable during the hydrate formation process. The thermostat bath was set to 274.15 K and kept stable throughout the experiment. MRI images were continuously obtained during the experiment.

2.3. Water flow process

There were sixteen experimental cases in this study. The water injection conditions and results for the sixteen cases are shown in Table 1. Case 1 was chosen to show the water flow process. After complete hydrate formation, the high-pressure vessel and backpressure pump were interconnected and the backpressure valve was set to 3200 kPa. During the backpressure adjustment, the backpressure was first set to 5000 kPa, and then it was set to 4000 kPa, and 3500 kPa. Finally, it was set to 3200 kPa and kept stable at that pressure for a period to ensure that there was no hydrate dissociation. The equilibrium pressure was 2980 kPa at a temperature of 274.15 K, as calculated by CSMHyd [4]. Thus, the hydrate remained stable and there was no dissociation during the changing of backpressure. Then, the water injection pump, which was set to the same pressure as the backpressure pump, was interconnected to the high-pressure vessel and the water temperature was set to 273.65 K to prevent hydrate dissociation. The hydrate-saturated sediment core was saturated with cooled deionized water injected at a velocity of 2 mL/min. Then, the cooled deionized water was continuously injected into the vessel with a velocity of 10 mL/min. Images were continuously obtained throughout the water flow process and the time interval was 2 min.