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Experimental investigation into formation/dissociation characteristics of methane hydrate in consolidated sediments with resistance measurement

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ABSTRACT

Natural gas hydrates (NGHs) are considered as a powerful potential energy resource. In this work, an experimental system containing a digital electrical bridge for resistance measurement was developed to gain an improved understanding of the formation/dissociation characteristics of NGHs in consolidated sediments. Using the apparatus, a dynamic methane hydrate formation method for sandstone cores was developed that can maintain a constant gas flow in the core during the methane hydrate formation process. As opposed to the traditional method that maintains a constant pressure in the core, higher resistance, permeability, and hydrate saturation for the core were obtained following methane hydrate formation equilibrium by using the proposed method. In order to investigate the methane hydrate formation characteristics in sandstone cores, the influences of core permeability, temperature, initial water saturation, and salinity on the resistance and hydrate saturation of two cores were analyzed in detail. The experimental results demonstrate that a relative lower temperature and higher initial water saturation in the core correspond to a higher hydrate saturation and water-to-hydrate conversation ratio. In the process of methane production from continent hydrate, the water output from the core with methane extraction was experimentally measured for the first time. It was found that, owing to the existence of free gas in the core holder, both the core resistance and methane production rate were not changed linearly with time. The methane production amount increased with an increase in initial water saturation, decrease in production pressure, or increase in pore volume in the core. It was demonstrated that water output is not a serious problem in the development of continental natural gas hydrate.

1. Introduction

Gas hydrates are inclusion compounds consisting of water and light gases under relatively low-temperature and high-pressure conditions. Natural gas hydrates (NGHs) are widely discovered in two different terrain environments: permafrost regions and offshore areas [1]. It has been estimated that the gross reserves of organic carbon in NGHs amount to approximately double those in conventional fossil fuels [2,3]. The manner in which to identify the distribution of NGH resources on earth effectively and perform further exploitation have attracted significant attention in the past decades. Meanwhile, as methane is the main gas component in NGHs, the pre-production of the NGHs can not only provide abundant methane resources but also prevent methane escaping into the air owing to climate change. Tested methane is a potent greenhouse gas that absorbs infrared radiation approximately 25 times more efficiently than carbon dioxide [4]. To date, several NGH development methods have been proposed that depend on laboratory works, such as (1) thermal stimulation [5,6], through increasing the deposit temperature to allow the hydrate to dissociate at a specified pressure; (2) depressurization [7,8], which decreases the deposit pressure below the hydrate formation pressure at a specified temperature; (3) chemical injection [9,10], whereby thermodynamic inhibitors such as alcohols are injected to shift the hydrate phase equilibrium conditions; and (4) CO_2 replacement [11–13], through injecting CO_2 (gas, liquid or emulsion) into NGH reservoirs to displace the methane trapped in the hydrate by forming new CO_2 hydrates. It should be noted that these techniques exhibit certain limitations, such as high heat loss in the thermal stimulation method [5,14,15], low injection rate and potential pollution when using chemical injection [16], and low reaction rate when using the CO_2 replacement method [17]. In contrast, although the ocean floor stability may be affected, depressurization is considered the most competitive method, owing to its high simplicity and energy efficiency [18].

In nature, the majority of NGHs are distributed in offshore area, which has attracted significant attention from researchers. This type of hydrate resource is usually found close to the sea floor, at water depths

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greater than 300 m, with the deposits being loosely unconsolidated sediments. Numerous laboratory experimental and simulation studies regarding the formation and dissociation behavior of oceanic NGHs have been reported in the literature. Chen et al. [19] systematically investigated the methane hydrate formation characteristics in coarse sands, silts, and natural clay marine sediments, they found that massive hydrate deposits were observed in the coarse sand and silt but not in the clay marine sediment. Li et al. [20] measured the change in electrical resistivity of hydrate-bearing sediments during the hydrate formation process. Based on the experimental results, the Archie equation was further used to describe the relationship between resistivity and hydrate saturation. In addition to electrical resistivity measurement, other technologies such as acoustic [21,22] and magnetic resonance imaging (MRI) [23,24] have been considered as alternative methods for studying the formation and distribution of NGHs in sediments. Wang et al. [25] investigated the synthetic effect of heat stimulation and depressurization on the dissociation of hydrate porous media, high gas recovery and thermal efficiency were obtained. Using the depressurization method, Yang et al. [26] studied the dissociation characteristics of two NGH deposit types, with one formed by means of excess gas and the other with excess water, during which MRI technology was used to monitor the liquid water distribution and quantify the methane hydrate amounts. Chen et al. [27] also investigated the basic dissociation behaviors of unconsolidated methane hydrate. Furthermore, they proposed a numerical simulation model for core-scale dissociation flow, and the simulation results agreed strongly with the experimental data. In addition to laboratory works, two typical oceanic hydrate exploitation field tests that have been performed are that in the Nankai Trough in Japan in March 2013 [28], and that in the Shenhu area of South China Sea in 2017 [29]. Both these tests used the depressurization method, and their gas production amounts reached $1.3 * 10^5$ and $\sim 30 * 10^5\ m^3$, respectively. It should be noted that, compared to low gas production quantity, these field trials involved significant capital investment. As proposed by the Department of Energy in the USA, the long-term research and development of gas hydrates should focus on demonstrating technologies that can achieve environmentally and economically feasible methane recovery [30].

In terms of continental hydrates, although certain deposits have been found globally, such as the Messoyakha field in the West Siberian Basin [31], Arctic regions of North America [32], Mackenzie Delta [33], Alaskan north slope [34,35], and Qilian mountain tundra of China [36,37], among others, owing to the significantly lower energy reserves in permafrost than that in the ocean, limited laboratory works regarding the formation and dissociation behavior of continental hydrates are available in the literature. Furthermore, compared to oceanic NGHs, the depths of continental hydrates usually range from 200 m to over 1700 m, with individual zones having a thickness of up to 100 m [38], which results in the deposits of the latter consisting more of consolidated sediments. For simplicity, Baldwin et al. [39] studied the formation and dissociation of THF hydrate in a Berea sandstone core with MRI, and stated that no significant differences were apparent between the characteristics of THF hydrate in sandstone and those in bulk. Stevens et al. [40] investigated the formation of methane and carbon dioxide hydrate in Bentheim sandstone samples, where the gas hydrate distributions in pores were characterized using MRI technology. Yousif et al. [41] conducted both experimental and simulation studies for recovering methane from Berea sandstone hydrate by means of depressurization, during which both the produced gas volume and hydrate interface position in the sandstone core were monitored based on MRI technology. Nazridoust et al. [42] used a computer modeling approach to study methane hydrate dissociation in a porous sandstone core, and stated that hydrate dissociation in a core was a sensitive function of the surrounding environment temperature, outlet pressure conditions, and permeability. It should be noted that the majority of these studies were not concerned with the water output accompanying gas extraction from the core. Moreover, the CO_2-CH_4 exchange method has been studied to produce methane from consolidated hydrates [43–46]. As reported by Birkedal et al. [44], 60% of methane in the sandstone hydrate could be recovered following the injection of CO_2. However, their experiments indicated that the action time when using the CO_2 replacement method is more than 600 h, even for a small core sample, which is significantly longer than when using the depressurization method.

It is generally believed that NGHs should be mined in the location where gas hydrates are initially formed. As stated previously, oceanic hydrate sediments are found in water that is usually deeper than 300 m, which complicates most current techniques for commercial production, and the reservoir properties of oceanic gas hydrate are generally unknown. In contrast, continental natural gas hydrate provides a superior opportunity for resource evaluation [38,40]. To date, several exploitation field tests for continental hydrate have also been performed; however, it should be noted that none of these tests have realized large-scale industrialized production [47]. The commercial application of continental hydrate energy needs further studies on the formation/dissociation characteristics of NGHs stored in sandstones. For these purposes, laboratory investigations provide greater time saving and energy efficiency than field tests.

The remainder of this paper is organized as follows. First, an experimental system is constructed for synthesizing a methane hydrate sample in sandstone cores and measuring the electrical resistivity variation in core samples during the hydrate formation and dissociation processes. Second, using the built apparatus, a dynamic natural gas hydrate formation method for sandstone cores is proposed, and the influence of temperature, core permeability, initial water saturation, and salinity on the resistance of two core samples and the hydrate saturation in these are investigated. Third, by using the depressurization method, the changes in core resistance, and gas production quantity and rate, during the hydrate dissociation processes from the core samples, as well as the water loss ratio of the core following methane hydrate dissociation equilibrium, are presented.

2. Experimental section

2.1. Experimental apparatus

An experimental system was designed and constructed for studying the natural gas hydrate formation/dissociation characteristics in sandstone cores by means of which the electrical resistance variations in moistened sandstone cores during both the gas hydrate formation and dissociation processes can be monitored in real time. The entire experimental system was placed in an air bath, which was used to provide a low environmental temperature for the reactor. A schematic of the experimental system is provided in Fig. 1, and it mainly consists of a core holder, feed gas injection auto pump, blind steel cell, and gas volume flowmeter. The core holder was a stainless steel cylinder with a maximum working pressure of 30 MPa and working volume of 500 ml, in which a rubber sleeve with an inner diameter of 2.5 cm was placed to contain the sandstone core. Two hollow steel columns with inner channel diameters of 0.15 cm were inserted into the holder from two sides to fix the core in the rubber sleeve and maintain a high pressure for the core. The channel in these two steel columns was used for methane flow into and outflow from the core. A digital electrical bridge with a two-electrode configuration, which were respectively placed at the end faces of the two steel columns, was used to measure the core resistance. The distance between the two electrodes was 5 cm. External pressure approximately 3 MPa greater than that within the core was maintained outside the rubber sleeve by means of a manual hydraulic pump. An auto pump connected to a high-pressure blind cell (1000 ml) was used to provide high-pressure methane gas to the moistened core for the gas hydrate formation therein. A back-pressure valve connected to the holder outlet line was used to set different methane hydrate production pressures. The methane gas produced from the core during