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Experimental study on the effect of methane hydrate decomposition on gas phase permeability of clayey sediments

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HIGHLIGHTS

- Gas phase permeability of clay decreases gradually with the hydrate decomposition.
- Permeability damage of clay with low saturation decrease after hydrate decomposition.
- Permeability damage of clay with high saturation increase after hydrate decomposition.
- Swelling of clay contribute to decrease of porosity after hydrate decomposition.

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ABSTRACT

Natural gas hydrates were widely distributed in marine sediments and permafrost areas, which have attracted global attentions as potential energy resources. Permeability characteristics of sediments determine the technical and economic feasibility of natural gas energy production and energy production efficiency from the hydrate reservoirs. But clay possesses significant water sensitivity and swelling characteristics in hydrate reservoirs, which may significantly affect permeability changes. Therefore, the mechanism significantly affects the gas energy production of hydrate reservoir. This study presented here focuses on the phenomenon of gas phase permeability changes due to hydrate decomposition. In this paper, the experimental study of the methane hydrate decomposition was carried out by depressurization, and the gas phase permeability characteristics changes of three kinds of clay before and after hydrate decomposition were investigated. The results show that the gas phase permeability of clay decreases gradually with the hydrate decomposition. The possible explanations for this phenomenon are that the formation of the bound water and swelling of clay, which block the pore channel of gas flow. In addition, after the hydrate complete decomposition, the value of the gas phase permeability damage (Ratio of permeability after hydrate decomposition to that before hydrate decomposition) of the clay firstly decreases and then increases with the increase of initial hydrate saturation. When hydrate saturation is 20%, Hydrate decomposition has the most influence on gas phase permeability damage of clay. And after the hydrate decomposition, the swelling of kaolin is less than illite and the swelling of illite is less than montmorillonite. The predicted porosity of clay after hydrate decomposition is calculated by Ives and Pienvichitr model and Tien's model. This work could be valuable to research on the gas energy production from the hydrate reservoir.

1. Introduction

Natural gas hydrate is a nonstoichiometric compound composed of a network of H_2O molecules that are hydrogen-bonded in a manner similar to ice and CH_4 guest molecules [1]. It has received wide attention due to its huge reserves as a future energy resource [2,3]. Hydrate production plays an important role in flow assurance, safety issues, energy recovery and climate change. Despite scientific interest in this compound and potential commercial energy production importance [4,5], many of the physical, chemical, and material properties of methane hydrate are as yet poorly constrained or unmeasured during the natural gas hydrate energy production [6–9]. During the hydrate energy production, The permeability of hydrate-bearing sediment is one of the most important parameters that determine gas and water flow processes and affect heat and mass transfer through hydrate sediments [10–13]. Previous studies have shown that in natural hydrate systems, permeability determines methane gas flux to the ocean [14], gas hydrate and free gas and water distribution [15,16], gas hydrate

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concentration and accumulation [17], and the efficiency in gas energy production from hydrate reservoirs [18–20]. Thus the permeability or hydraulic conductivity of sediments with hydrate is necessary and significant to the energy production of the gas hydrate resources, which affects the production efficiency of natural gas energy from hydrate reservoirs [21,22].

But, natural gas hydrate reservoirs have unique characteristics in the South China Sea [23]. In previous investigations, it was found that natural gas hydrates are mainly transported to stable regions and filled in cracks from high flux methane gas through structural migration channels [24]. And there are specimens with excess methane gas in situ subsea that are analogs for natural gas-rich systems such as those that may occur when gas recycles into an upward-migrating base of hydrate stability [25,26], which is gas-saturated sediments containing methane hydrate. In 2007, gas hydrate cores were obtained through drilling exploration in Shenhu area of South China Sea and related researches have shown most local methane hydrates producing formations contain large amounts of clay minerals that were originally deposited during sedimentation (detrital clay), or precipitated from fluids flowing through the matrix (authigenic clay) [27,28]. In addition, previous studies have found that the dissolution of hydrates causes swelling and shrinkage behaviors of clay, which results in natural disasters such as submarine landslides [29]. So, compared with sands, authigenic and detrital clays can cause loss of permeability by unique water sensitivity and swelling mechanisms, which is one of the critical geomechanical parameters that influence the energy production potential of natural gas from hydrate reservoirs [28] (Water molecules overcome the gravitational force of aqueous solution and wedge into the inner layer of clay particles, becoming the bound water, which causes clay particles to swell [27,28]. Two types of mechanisms for clay swelling were identified: crystalline swelling and osmotic swelling). Clay swelling has been long recognized as one of the major causes for formation damage in hydrocarbon reservoirs. From the energy-production perspective, the mechanism may induce damage of the energy production efficiency for the hydrate reservoirs. So it is very meaningful to study the permeability damage of the sediments caused by the above mechanism for the hydrate energy production.

However, very few permeability experiments for methane hydrate-bearing clayey sediments with different hydrate saturations or hydrate decomposition in the laboratory are available. And the research on the effects of the above mechanisms on hydrate energy production is extremely scarce. Zhao et al. assessed the change of natural gas production process for natural gas hydrate using depressurization [11]. Li et al. experimentally measured the absolute permeability and the water effective permeability with fluid water under serials of hydrate saturations (0–31% in volume) [30]. Sakamoto et al. measured the water permeability of methane hydrate-bearing sands with hydrate decomposition by nitrogen-water simultaneous injection and normal hot-water injection. The results indicated that water permeability decreased with methane hydrate reformation and growth and water permeability increased with methane hydrate decomposition in the hydrate-bearing sands [31]. Wang et al. analyzed scaling criteria for methane hydrate dissociation in sediment using the decompression method [22]. Wang et al. analyzed gas recovery from methane hydrate deposits and the concomitant ice generation by using depressurization mode [32]. In addition, the researchers also studied permeability of clayey hydrate reservoirs. Oyama et al. used a natural core obtained from the eastern Nankai Trough to perform hydrate dissociation experiments. The results presented the hydrate dissociation characteristics of low permeability methane-hydrate-bearing cores are different from that of a high-permeability artificial core [33]. Okwannanke et al. carried out the gas phase permeability experiments using methane hydrate sediments and obtained a small amount of permeability data, discovering the change law of gas phase permeability of gas hydrate-bearing clayey sediments is different from that of artificial gas hydrates-bearing sands [34]. Oyama et al. carried out the mud and floc erosion experiment in Japan's offshore turbidite sediments. From these experiments, it is considered that mud and floc erosion have an insignificant effect on gas production from methane hydrate reservoir [35].

Due to the effect of clay surface electric field force, water molecules produced by hydrate decomposition will cause the volume of pore space reduce by swelling of the clay [36,37]. But the mechanisms by which these changes occur for the damage of hydrate energy production remain largely unresolved [38]. In the absence of sufficient experiment data, the hydrate reservoir permeability damage caused by the various kinds of clay swelling is difficult to be predicted. We focused on the effect of the unique mechanism of clay on the permeability of hydrate reservoirs during the hydrate decomposition. This study investigated the relationship between gas phase permeability and hydrate dissociation in kaolin, montmorillonite and illite and effect of swelling of clay on gas flow, which is valuable for the economic feasibility and production efficiency of hydrate energy production in the South China Sea.

2. Experimental methods

2.1. Experimental apparatus

Fig. 1 shows a schematic diagram of the experimental device adopted in this study for the effective gas phase permeability measurements during hydrate stabilization and decomposition in the hydrate-bearing clayey sediments. In this study, experimental device can simulate the *in situ* temperature, pore pressure and overburden pressure conditions of hydrate formation. The reaction chamber is typically 40 mm in diameter and 215 mm in length with pressure capacity of 20 MPa. The axial pressure is exerted by an axial compression pump which is connected to the piston at one end of the reaction chamber, which simulates the overburden pressure (with pressure capacity of 40 MPa and volume of 200 ml). The temperature ranges from −30 °C to 90 °C with a controlling precision is 0.05–0.1 °C, which were controlled by the water bath (model XT5704LT-R30). The pore pressure of the hydrate-bearing sediments is supplied and controlled by an ISCO 260D pump. The differential pressure between the inlet and outlet of the reaction chamber is measured by a differential pressure sensor (ranges from 0 to 1 MPa, with an accuracy of ± 0.01 kPa), and the data can be transmitted to the computer through the AD module and automatically recorded and analyzed. The steady-state flow is controlled by a flow controller (The flow range is 0–100 ml under atmospheric pressure, and the accuracy is ± 0.5%) which is connected to the outlet end of the system.

2.2. Materials

In this study, the effect of hydrate decomposition on gas phase permeability of clay was investigated. Kaolin, montmorillonite and illite were used in the experiments in order to compare effect of different

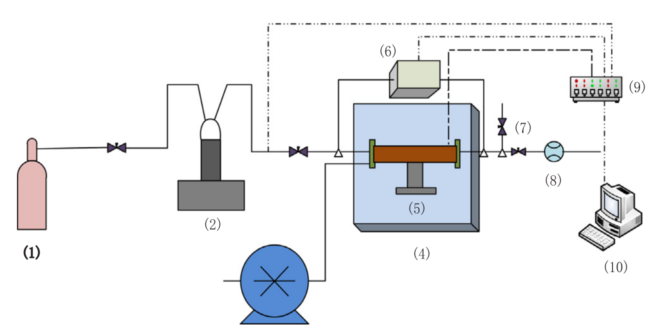

Fig. 1. Schematic of the experimental device system. (1) CH_4 gas, (2) ISCO pump, (3) axial compression pump, (4) Glycol water bath, (5) reaction chamber, (6) differential pressure transducer, (7) exhaust, (8) flow controller, (9) A/D Module, (10) computer.