Research paper

Experimental study on the gas phase permeability of montmorillonite sediments in the presence of hydrates

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ABSTRACT

Permeability is a key parameter influencing fluids flow capacity and the gas production rate of the marine hydrate-bearing sediments. In this study, a series of experiments were carried out to find out the dependence of the gas phase permeability of montmorillonite on different hydrate saturations at effective axial stresses of 1 MPa, 3 MPa. And an experimental study of the decomposition of methane hydrate (MH) by depressurization was carried out. It is found that with the increase of gas hydrate saturation, the gas phase permeability of montmorillonite shows a trend of decreasing first and then increasing, due to the effect of blockage of hydrate particles and the increase of pores in inter-aggregate zones during methane hydrate formation. Furthermore, the gas phase permeability of clay clearly decreases gradually with the increasing decomposition time under different hydrate saturations, due to the formation of the bound water and the swelling of clay. The magnitude of CH_4 coefficient of permeability K_a and that of transmission D^* reach 10^{-7} and 10^{-8}, respectively. K_a is also known as hydraulic conductivity in the Darcy law. It is defined as the unit flow rate under the unit hydraulic gradient, indicating the difficulty of the fluid passing through the pore skeleton. And D^* is the coefficient of transmission based on a modified form of Fick's law, which is a function of the volume-mass properties of the soil and the gas density. After the complete decomposition of the hydrate, the permeability coefficient K_a decreases with the increase of initial hydrate saturation when $4.02\% \leq S_h \leq 15.15\%$. However, $15.15\% \leq S_h \leq 27.26\%$, the values of K_a increase with the increase of initial hydrate saturation. These experiments helped us understand some important aspects of MH behavior such as saturation of hydrate formation, effective stress, and permeability change during decomposition. This work could be useful to research on the production efficiency of marine gas hydrate reservoir.

1. Introduction

Natural gas hydrates (NGH) are ice-like crystalline solid compounds in which a water molecule structure contains embedded methane molecules under low-temperature and high-pressure conditions (Sakamoto et al., 2010). When $1\,m^3$ of NGH is decomposed, approximately $164\,m^3$ of methane gas is produced at standard conditions (Sloan, 1998). Through scientific drilling projects and field-scale research projects, many countries have confirmed the widespread distribution of gas hydrates in marine sediments and permafrost areas where the geologic conditions are favorable for their formation and stability (Li et al., 2013). To meet the increasing global energy demand, NGH have attracted much attention to be investigated widely (from laboratory to field) in the world as a potential energy resource (Song et al., 2013; Yang et al., 2014).

There is still much uncertainty in determining NGH's quantities and distribution in nature. But, two critical conclusions can be formulated:

(1) hydrates likely exceed the total energy content of 'conventional' sources of natural gas (Kvenvolden, 1993) and (2) NGH predominantly exist in offshore environments and onshore permafrost (Collett, 2002; Zhao et al., 2013). Irrational exploitation of NGH may lead to serious environmental problems and climate disasters (Kvenvolden, 1988). The rational exploitation and effective utilization are the keys to the production of natural gas from hydrates. Production of natural gas from hydrates can be achieved by inducing decomposition through depressurization (Ji et al., 2001; Song et al., 2015; Zhao et al., 2015; Liu et al., 2014), thermal stimulation (Mcguire, 1982), inhibitor injection (Kamath and Godbole, 1987), or their combinations (Moridis, 2003). All are based on the in-situ decomposition of NGH that is transformed into methane gas and water. The process must be related to the flow of gas and water in the reservoir, and the gas/water phase permeability of the reservoir are the important factors to evaluate the gas production efficiency (Minagawa et al., 2007; Zhao et al., 2011a,b). The effective permeability (k_{eff}) of each phase in the hydrate-bearing sediments

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reflects the ability of fluids to migrate in the porous media and also affects the heat and mass transfer rates among different phases (gas, water, hydrate, and sediment grain) (Zhao et al., 2014; Song et al., 2016). However, the permeability change characteristics of the gas hydrate reservoir are complicated which will be affected by the hydrate saturation, the porosity, the properties of sediment particles, and so on (Moridis, 2014; Zhao et al., 2011a,b; Wang et al., 2016).

In order to safely and efficiently exploit NGH reservoirs and predict permeability change characteristics of the NGH reservoirs, researchers proposed various flow models to describe the mechanism of infiltration in sand and clay (Delli and Grozic, 2013; Holder and Angert, 1982; Jeannin et al., 2002; Masuda et al., 1997). And many researchers have investigated the flow properties of the porous media in the process of stable presence of hydrate and hydrate decomposition using different methods. Delli and Grozic (2014) performed a series of experiments to investigate the variation of permeability in gas hydrates-bearing sediments where carbon dioxide hydrates were formed. Sakamoto et al. (2010) carried out the hydrate decomposition experiment by injecting hot water, the results indicated that water phase permeability change characteristics were influenced by the decomposition of hydrate. Li et al. (2013) found that water phase permeability changed with the different water injection rates in the dry quartz sands due to the hydrate dissolution. Kumar et al. (2010) simulated the change of permeability in the process of hydrate decomposition using a 1-D numerical model. These studies were focused on the hydrate-bearing sands, and only studied the effect of hydrate saturation on permeability in the process of stable presence of hydrate and the hydrate decomposition. It can be found from the above research, few studies were conducted on hydrate-bearing clay, which may present a significantly different permeability behavior during the hydrate decomposition. In the previous study, it was found that the change of clay structure in the process of hydrate formation resulted in the change of permeability of clay (Liu et al., 2016). However, due to the ion action, the water produced during the decomposition of the hydrate will react with clay particles strongly (Chang et al., 2009; Dékány, 2002; Klapproth et al., 2007), which will affect the permeability behavior of the porous medium.

In 2007, gas hydrate cores were obtained successfully through drilling exploration in Shenhu area of South China Sea, and gas hydrate saturation is 20%–40% in the exploration zones (Zhang et al., 2007). The gas hydrate cores drilled from SH-2, SH-3 and SH-7 site are mainly composed of terrigenous minerals, clay mineral and biogenic carbonate, and the main ingredients of the drilled cores are illite, montmorillonite and kaolin clay (Zhang et al., 2007). Montmorillonite used in this study is very similar to the material compos of SH's cores. Therefore, the montmorillonite hydrate-bearing sediments with different hydrate saturations were used to determine the gas phase permeability by a series of laboratory-scale experiments and observe the gas phase permeability change characteristics during the hydrate decomposition using the depressurization method.

2. Experimental methods

### 2.1. Experimental apparatus

Fig. 1 shows a schematic diagram of the experimental device that was adopted in this study for the effective gas phase permeability measurements during hydrate stabilization and hydrate decomposition process in the hydrate-bearing sediments. It can simulate the *in situ* pore pressure, temperature and stress conditions of hydrate formation in a cylindrical reaction chamber. The reaction chamber is typically 40 mm in diameter by 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. The temperature ranges from −30 °C to 90 °C (The temperature controlling precision is 0.05 °C–0.1 °C), which can be controlled using the water bath (model XT5704LT-R30). The pore pressure of the hydrate-bearing sediments is supplied and controlled by an ISCO 260D pump. The pressure difference between the inlet and outlet of the reaction chamber is measured by a differential pressure sensor (ranges from 0 to 1 MPa), and the data can be recorded and analyzed by the computer automatically. 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. Clay type

A few number of gas hydrate-bearing cores have been obtained in the Shenhu area of the South China Sea (Liu et al., 2012). The main components of the sediments are clay and silt sand. But the amount of ocean soil cores is not enough to support a complete experiment. In this study, the montmorillonite was chosen to conduct the permeability experiment as both the composition and the grain size distribution of marine soil sediments are similar to that of montmorillonite. Table 1 shows the comparison of montmorillonite and marine soil in the grain

Table 1
Comparison of the grain size between montmorillonite and Marine Soil.

Grain Size	< 4μm	4μm–63μm	> 63μm
Montmorillonite	29.47%	67.46%	4.07%
Marine Soil	28.74%	67.043%	4.217%
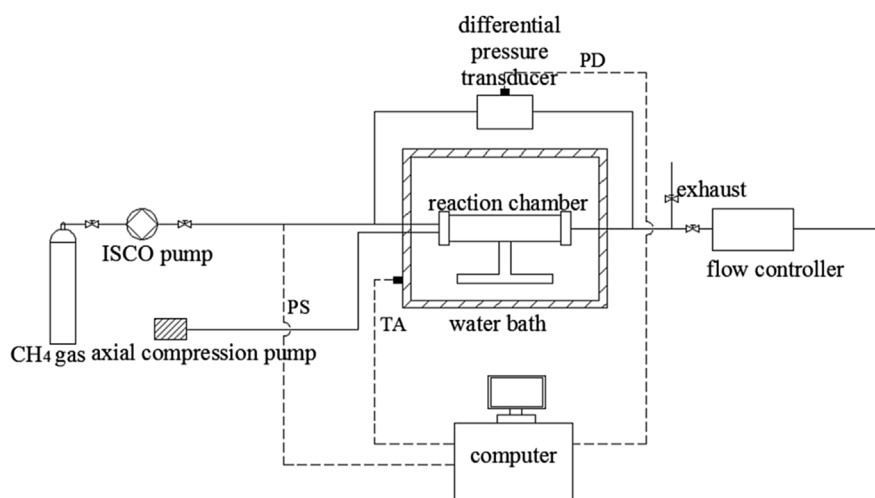
Fig. 1. Schematic of the experimental device system.