Journal of Natural Gas Science and Engineering 36 (2016) 378–384

Contents lists available at ScienceDirect

Journal of Natural Gas Science and Engineering

journal homepage: www.elsevier.com/locate/jngse

Experimental study on the gas phase permeability of methane hydrate-bearing clayey sediments

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ARTICLE INFO

Article history:
Received 2 July 2016
Received in revised form
16 September 2016
Accepted 24 October 2016
Available online 26 October 2016

Keywords:
Hydrate-bearing sediment
Gas phase permeability
Hydrate saturation
Effective axial stress

ABSTRACT

The permeability of hydrate-bearing sediment is one of the important parameters that influences the gas production rate of the hydrate reservoirs. In this study, a series of experiments were performed to investigate the gas phase permeability of kaolin clay with different hydrate saturations (0%, 2.59%, 4.23%, 11.44%, 16.84%, 22.91%, 27.57%, 28.85%, 34.16%, and 40.46%) at effective axial stresses of 1 MPa, 3 MPa. The results indicated that the gas phase permeability of the kaolin clay firstly decreases and then increases with the increase of hydrate saturation, due to the effect of blockage of hydrate particles and the increase of pores in inter-aggregate zones during methane hydrate formation. And there is a critical hydrate saturation that the gas phase permeability of hydrate-bearing clayey specimen equals to that of pure clayey specimen. Furthermore, the gas phase permeability of hydrate-bearing clayey specimens under high effective axial stresses is less than that under low effective axial stresses, due to the compaction of pore spaces and pore throats. And the stress sensitive index clearly increases with the increase of hydrate saturation, and the increase tendency become gentle when the hydrate saturation higher than 30%.

1. Introduction

As the rapid development of global economy and the expanding desire for energy, the energy shortage problem has become a strategic challenge for national economic development. Gas hydrate is an ice-like crystalline solid compound composed of water and guest molecules, such as methane, and commonly found in permafrost regions and outer continental (and insular) margins (Booth et al., 1998; Ruppel and Noserale, 2012; Sloan and Koh, 2008). It has received continuous attention worldwide due to its great utilization value as a future energy resource (Lu et al., 2007; Udachin et al., 2007). However, the exploitation of gas hydrate may cause marine geological disasters and global climate change which has obtained increasing attention around the world (Delli and Grozic, 2014; Goel, 2006; Grozic, 2010; Lashof and Ahuja, 1990). There are about four kinds of extraction methods for gas hydrate, such as depressurization method (Ji et al., 2001; Sloan and Koh, 2008), thermal stimulation method (McGuire, 1981), inhibitor injection method (Kamath and Godbole, 1987) and combined exploitation method (Lee and Holder, 2001; Moridis, 2002). The feasibility of these extraction methods depends on the specific conditions of gas hydrate-bearing layers, which are uncertain during the hydrate production. Permeability is one of the critical performance parameters used in the simulation of gas hydrate production, which determines the ability of gas or fluid flow in porous media and heat/mass transfer among these phases (Li et al., 2013). The permeability of gas hydrate-bearing sediments is critically affected by the presence of hydrate within porous media, which should be fully investigated to assess the gas productivity and the feasibility of the extraction method for gas hydrate.

In order to describe the mechanism of infiltration in sand and clay, researchers proposed various flow models to explain the physical meaning of the permeability coefficient K under the assumption of laminar flow (Delli and Grozic, 2013, 2014; Masuda et al., 1997; Scheidegger, 1958; Spangenberg, 2001). And many researchers have carried out experimental studies on the flow properties of gas and water in the hydrate-bearing sediments. Scheidegger (1958) and Moridis (2014) considered that the permeability of porous media depended on the sand particle size, porosity, and hydrate saturation. Minagawa et al. (2005) measured the water permeability of methane hydrate-bearing sediments, the results indicated that water permeability significantly depended on the hydrate distribution in pores related to grain size. Kumar et al. (2010) performed a series of experiments to measure the gas phase

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http://dx.doi.org/10.1016/j.jngse.2016.10.055
1875-5100/

permeability at varying CO_2 hydrate saturations in the porous medium made of packed glass beads, and found that the permeability decreased with the increase of hydrate saturation. Sakamoto et al. (2010) investigated the permeability change behaviors during hydrate dissociation by hot-water injection and depressurization. These studies were focused on the hydrate-bearing sands, and few studies were conducted on hydrate-bearing clay, which may present a significantly different permeability behavior. The specific surface area of clay particles is larger than that of sand particles, and clay particles always aggregate into clusters, rather than take the form of a single clay particle, the negative charges on the surface of the clay particles and the polarity of water molecules attract each other due to the effect of surface force (Chang et al., 2009; Marion et al., 1992), which will affect the permeability behavior of the porous medium.

In 2007, gas hydrate cores were obtained through drilling exploration in Shenhu area of South China Sea, and hydrate saturation of 20%–40% was observed in fine grained sediments in this area (Liu et al., 2012). The gas hydrate cores drilled from SH2B and SH7B site are mainly composed of terrigenous minerals, clay mineral and biogenic carbonate (Liu et al., 2012), and the main ingredients of the drilled cores are illite, chlorite and kaolin clay (Liu et al., 2010). According to the literature (Wu et al., 2008), the hydrate cores are drilled from ~225 m below the seafloor, where the effective axial/formation stress is about ~3 MPa. And the permeability varies under different effectvie axial/formation stresses. Therefore, it is necessary to measure the permeability of clay in the presence of gas hydrate under different effective axial/formation stresses.

In this study, a series of laboratory-scale experiments were conducted to investigate the gas phase permeability of kaolin clay with different hydrate saturations at different effective axial stresses.

2. Experimental methods

2.1. Experimental apparatus

The schematic of the permeability measurement system is shown in Fig. 1. It can simulate the *in situ* pressure, temperature and stress conditions 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 stress is provided by an axial compression pump (with pressure capacity of 40 MPa, and volume of 200 ml). The temperature ranges from −20 °C to 50 °C, which can be adjusted by the air bath (model GDW-010 L) with dimensions of 1000 mm (L) × 1000 mm (W) × 1000 mm (H). The steady-state flow and pressure are 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 550 kPa, with an accuracy of ±0.12 kPa). A mass flow meter and a temperature sensor are equipped to obtain the gas flow and temperature respectively, and these data can be recorded and analyzed by the computer automatically.

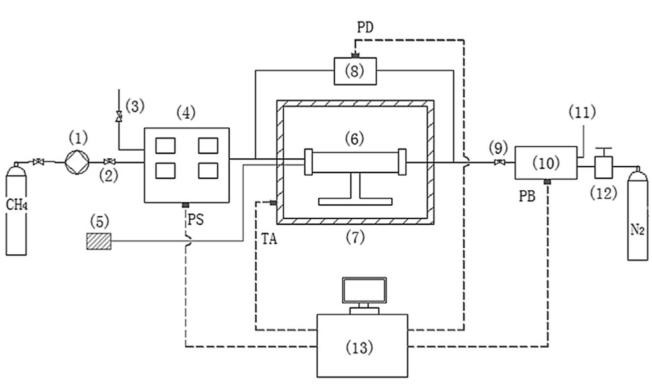

Fig. 1. Schematic of the permeability measurement system.

(1) ISCO pump, (2) intake valve, (3) vent valve, (4) data board, (5) axial compression pump, (6) reaction chamber, (7) air bath, (8) differential pressure transducer, (9) outlet valve, (10) back pressure regulator, (11) exhaust, (12) pressure regulator, (13) computer

2.2. Clay type

A few number of gas hydrate-bearing cores have been obtained in the Pearl River Mouth Basin, offshore China. The sediments are mainly composed of clay and silt sand (Chen et al., 2011; Liu et al., 2012). The composition and the grain size distribution of these sediments are similar to that of kaolin clay. In this study, the kaolin clay was chosen to simulate the marine sediments to do the permeability experiment. Table 1 and Fig. 2 show the major components and grain size distribution of kaolin clay, respectively.

2.3. Experimental procedure

In this study, the specimen was prepared in the reaction chamber by mixing the cooled kaolin clay and ice powders, the methane hydrate was generated by injecting methane gas into the reaction chamber under above zero conditions. Firstly, the reaction chamber and the pipelines were dried by flowing methane gas at a low flow rate. Meanwhile, ice powders were manufactured by using an ice crusher to break the prepared freezing distilled water, and then screened by a standard 60-mesh sieve. To obtain the methane hydrate-bearing specimens with 45% porosity and designated hydrate saturations, 386.25 g of kaolin clay and a known amount of ice powders (according to the initial ice saturations S_i of 5%, 10%, 15%, 20%, 25%, 30%, 35%, 40%, 45%) were evenly mixed and filled in the reaction chamber. All the above experiment processes were performed in the cold storage with a temperature of −10 °C. Secondly, the reaction chamber was removed from the cold storage and connected into the permeability measurement system. Then the methane hydrate was generated by injecting CH_4 with a constant pressure of 5.5 MPa under a temperature of 1 °C. During the hydrate formation, the pressure and the temperature were maintained by the ISCO pump and air bath respectively. When the gas consumption did not change after several hours as indicated by the display of the ISCO pump, it can be considered that the methane hydrate had been fully generated. Finally, the gas permeability experiments were conducted. The back pressure regulator was set to 5.3 MPa and axial loads of 6.3 MPa and 8.3 MPa were applied to simulate the formation stresses *in situ* by the axial compression pump (corresponding to formation stress/effective axial stress σ' of 1 MPa and 3 MPa). A constant CH_4 gas flow was supplied by the ISCO pump, and the pressure difference across the reaction chamber was measured by using the differential pressure transducer.

In this study, the axial loads were determined by the effective stress principle (Kim et al., 1993), as shown below:

Table 1
Major components of kaolin clay.

Component	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	MnO	K_2O	Else
Content/%	52	35	1.5	2.6	1.0	0.1	0.13	1.3	6.37