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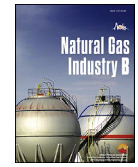

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Research Article

Permeability experiments on the methane hydrate in quartz sands and its model verification☆,☆☆

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Abstract

The permeability of porous media is an important physical parameter that affects the exploitation of natural gas hydrate. At present, however, the measurement methods and porous media used for investigating the permeability are so different that neither well-recognized experiment methods nor measurement results are available yet. In this paper, a one-dimensional test apparatus was developed to measure the permeability of porous media with methane hydrate. By virtue of this apparatus, methane hydrate was generated and the flow rate and pressure difference of inflow liquid water were measured in a stable flow pattern. Then, based on the basic principle of the Darcy's Law, the permeability of 30–40 mesh quartz sands with a methane hydrate system was calculated using steady-state water injection. And the experimental results were obtained. First, this apparatus can provide the flow rates and pressure differences of stable fluid under constant pressures and temperatures, so it satisfies the basic conditions of the Darcy's Law and consequently the permeability is calculated. Second, with this apparatus, the methane hydrate saturation in permeability experiments can be controlled effectively, so that the reliability and repeatability of permeability measurement of methane hydrate bearing quartz sands are ensured. Third, methane hydrate crystals are formed and gradually grow in the pore center, occupying the pore space and blocking the flow channel. And the liquid effective permeability decreases sharply with the increase of methane hydrate saturation. Fourth, the permeability calculated by the Masuda, Dai and Li models is 13.0, 7.0 and 4.0, respectively and the calculated values are in accordance with the experimental results. These research results provide experimental data and a theoretical calculation basis for the quantification of fluid permeability of methane hydrate bearing porous media.

Keywords: Porous media; Quartz sand; Methane hydrate; Permeability; Saturation; Experimental simulation; Mathematical model; Verification

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1. Introduction

Natural gas hydrate (NGH) is dominated by methane hydrate (MH). NGH is widely distributed in the nature and usually found in deep sea and permafrost regions [1,2]. It is considered as a great potential alternative energy resource, and has attracted the attention of scholars from all over the world [3,4]. NGH is often decomposed via depressurization [5,6], thermal stimulation [7−9], chemical method [10−12], and replacement with CO_2 [13,14], when its thermodynamic equilibrium is broken. In the NGH exploitation process, the permeability of the sediment system is a very important basic physical parameter. The sediment particle size, pore size distribution, specific surface area and saturation distribution of solid MH all have a great effect on the permeability. For the specific sediment pore structure [15], the saturation of MH is critical, so its quantification is meaningful to the basic research and exploitation engineering of NGH.

Scholars home and abroad have studied the permeability of the porous media with methane hydrate experimentally and theoretically. Direct measurement by experiments is the basic method and important basis for obtaining the permeability of different porous media with methane hydrate. The measurement methods include the un non-steady state method and the steady state method. As to the non-steady state method, the permeability is calculated by calculus, since the temperature, pressure and pressure difference all vary with time when fluid flows in a pore structure. However, the non-steady state method is rarely reported. Jaiswal et al. [16] investigated the relationship between the effective permeability and the gas−liquid relative permeability of the sediment containing methane hydrate and the MH saturation by the experiment of JBN non-steadystate method and found that the relative permeability in the unsteady flow state is mainly affected by pore distribution in a sediment and the MH saturation in pores. As to the steady state method, the permeability is calculated according to the Darcy's law by measuring the pressure difference at both ends of a pore structure and the volume flow rate when fluid (gas or liquid) flows through the pore structure of porous media in the condition of stable seepage. The steady state method is simple, and can provide stable and consistent measurement results. Therefore, this method is adopted in most experimental studies.

In terms of mathematical models, Delli et al. [17] used the Ottawa 20/30 silica sand with particle size of 600−852 μm (or 720 μm averagely) to simulate the porous media, and synthesized CO_2 hydrate with different saturations by the partial water saturating method, with the MH saturation as high as 45%, and then measured the effective permeability of the liquid water in the steady state. Based on the experimental data, and considering two MH distribution types (particle trapping and pore filling), the mixed weighting model of permeability of the porous media with methane hydrate was put forward, in which the weight coefficient was related to the MH saturation. Song Yongchen et al. [18] simulated the porous media using the series of glass sands produced by the AS-ONE company of Japan and measured the influence of methane hydrate on porous media permeability with distilled water. He found an exponential relationship between the permeability and the MH saturation, and obtained the permeability model of glass sands by fitting experiment results; however, this model is only applicable to BZ-01 and BZ-02 glass sands, and has not been verified with other glass sands. Masuda et al. [19,20] put forward a mathematical model to describe the relationship between the absolute permeability or relative permeability of porous media with methane hydrate and the MH saturation by assuming the porous media as capillary and the methane hydrate generated on the inner wall of the capillary. In this mathematical model, the permeability decline index (N) depends on the pore structure of porous media, with the value changing between 1 and 25. However, the physical interpretation and basis of N values were not illuminated. Dai and Seol et al. [21] established the linear relationship between the product of the tortuosity and the specific surface area and the MH saturation, and then put forward a pore network model to describe the relationship between the relative permeability of water and the MH saturation and identify the impact of MH storage state and heterogeneity in the pore on the permeability. The simulation results show that the increase of MH content in sediment can lead to the decrease of porosity and the increase of tortuosity. This model adopts simple parameters, and provides the calculation results in good agreement with the experiment results. However, the selection of the product parameters depends on the experimental data, and as for the specific porous media, they need to be determined by experimental measurement.

In terms of experimental studies, Kleinberg et al. [22] measured the permeability of Berea sandstone with methane hydrate by the NMR method, and calculated the relative permeability of water using the Kenyon formula which is widely used in nuclear magnetic resonance (NMR) logging. According to the permeability measurement of No.5 Berea sandstone, the methane hydrate is formed in the center of the sandstone pore rather than on the surface. Johnson et al. [23] measured the gas−liquid relative permeability of the sediment samples from the Albert Mt. in the north slope of Alaska, the US, with the particle size of 31−125 μm. They used nitrogen to measure in the conditions of steady flow rate and pressure difference. The results show that methane hydrate (the saturation of 1.5−36%) can significantly reduce the permeability of porous media. Kumar et al. [24] conducted the experiment to measure the permeability of the porous media with methane hydrate using CO_2 and glass beads with particle size of 88.90−149.86 μm, through stable injection of CO_2. The results show that the absolute permeability of the porous media without methane hydrate is 66.9 D. They compared the experiment results with the Masuda model and found that when the MH saturation is less than 35%, the methane hydrate tends to generate on the particle surface, and the results of Masuda model agreed with the experiment results when the permeability decline index N is 3. Moreover, as the MH saturation increases, N rises. When the MH saturation is 42% and 49%, N is respectively equal to 4 and 5. The methane