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Experimental measurement and mathematical model of permeability with methane hydrate in quartz sands



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HIGHLIGHTS

• We develop an apparatus for hydrate formation and permeability measurement with water.

- We measure water permeability with methane hydrate in three different quartz sands.
- Permeability features are different with hydrate saturation higher or lower than 10%.
- Simple cubic sphere pack with uniform grain diameter was used in the model.

• We develop a new relationship between water permeability and hydrate saturation.

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ABSTRACT

Natural gas hydrates widely distributed in marine sediments and permafrost areas have attracted global attentions as potential energy resources. The permeability of sediments with or without hydrate is an essential and critical parameter that could determine the technical and economical feasibility of gas recovery from hydrate reservoirs. The saturation of hydrate in the solid phase significantly affects the pore size, the pore volume, the distribution of reservoir pore throat size, etc., which are key factors determining the permeability of the hydrate-bearing deposit. In this study, the absolute permeability and the water effective permeability were experimentally measured with fluid water under a serials of hydrate saturations (0-31% in volume). Hydrate saturations were controlled and calculated precisely based on the amount of injected and produced gas/water, and the system pressure and temperature. Unconsolidated quartz sands with different particle size (200-300, 300-450, 450-600 µm) were used as the porous media. The absolute permeabilities of the above quartz sands were 21.11, 35.53 and 52.32 Darcies, respectively. The experimental results indicated that the characteristics of the permeability were different with the hydrate saturation lower and higher than 10%. When the hydrate saturation increased from 0 to 10%, there was a sudden drop for the permeability, which indicated that the appearance and the existence of the solid hydrate phase in the porous media affected the permeability significantly. On the other hand, this effect lightened when the hydrate saturation higher than 10%. With different hydrate crystal growth habit, a new relationship between the ratio of the permeability in the presence and the absence of hydrate and the hydrate saturation was developed. Two patterns of the pore filling models with the hydrate saturation lower and higher than 10% were used to fit the measured experimental data. The overall relationship and the values of the saturation exponent were continuous and consistent with hydrate saturation lower than 31%.

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1. Introduction

Natural gas hydrates (NGHs) are non-stoichiometric crystalline solids formed by water molecules and small gas molecules (e.g., methane, ethane and carbon dioxide, etc.) at high pressure and low temperature [1-3]. NGHs are widely distributed in marine



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sediments and permafrost areas where the geologic conditions are favorable [4–6]. NGHs are considered to be one of the most potential energy resources and have attracted global attentions [7,8]. The estimates of the naturally-occurring gas hydrates continue to range over several orders of magnitude. Boswell and Collett [9] reviewed the gas hydrate resource assessments, and pointed out two types of Technically Recoverable Resources (TRR) of concentrated gas hydrate accumulations: (1) grain-displacing hydrate in the form of veins, nodules and fracture-fills, (2) pore-filling hydrate in sands. As yet, there have been no field tests or published numerical modeling studies that indicated feasible recovery of gas from the grain-displacing hydrate, which was generally caused by the lack of matrix permeability and low gas flow rates. On the other hand. NGHs within sand reservoirs are considered to be potentially producible in a manner that is consistent with commercial production and using current technologies. Field tests include Mallik 2007/2008 [10]. Gulf of Mexico 2009 [11.12] and Nankai Trough 2013 [13] have documented the gas production potential in the hydrate-bearing sand reservoirs. The essential difference between the above two types is the intrinsic permeability of the sediments (prior to the formation of the gas hydrate), which controls the gas and water mobility and the ability to transmit pressure and temperature within the reservoirs. Hence, investigations on the permeability of the sediments with or without hydrate are necessary and significant to the economical recovery of the gas hydrate resources.

Current laboratory researches on gas hydrates include the rapid hydrate formation [14] and several hydrate dissociation methods, e.g. depressurization [15–18], thermal stimulation [19,20], thermodynamic inhibition [21,22], and CO_2 replacement [23,24]. There are some joint methods based on the above single principles using complex vertical and/or horizontal well patterns, such as the inverted five-spot water flooding method [25], the huff and puff method [26,27], the steam assisted gravity drainage (SAGD) [28], and other combined methods [29].

For the above hydrate dissociation methods, the processes of the natural gas been produced from the hydrate-bearing deposits all refer to fluid flow and transport in the porous media. The permeability of porous media controls the flow rate of fluid (gas and water) [8,9]. Furthermore, the permeability affects the pressure, heat and chemical transfer among gas, water, hydrate, and sediment grain via fluid migration [30]. Yin et al. [31] reviewed the gas hydrate dissociation kinetic models for NGHs as a potential energy resource. They summarized the kinetic models thoroughly and confirmed that the permeability had significant effect on hydrate dissociation and gas production from NGH reservoirs. The presence of gas hydrate in porous media reduces the permeability, which is one of the critical geomechanical parameters that influence the production potential of natural gas from hydrate reservoirs [16,26,32]. From the gas-production perspective, a hydrate-bearing reservoir in coarse porous media (rocks or sandstones with large particle size and pore space, and less cementing effect between grain particles) with high permeability is a much desirable gas source than that in fine sediments (silt and clay, etc.) with low permeability. For the field test and commercial application of the gas production from hydrate reservoirs in the marine sediments and permafrost area, it is significant and meaningful to understand how gas hydrate in the pore space affects permeability. The prediction accuracy of the gas production process and the economic viability depends strongly on the hydrate distribution and its effect on permeability in the sediments.

In general, there are several methods to measure the permeability, including the nuclear magnetic resonance (NMR), X-ray computed tomography (CT) and the water or gas injection methods, etc. Kleinberg et al. [33] measured the methane hydrate-bearing sandstones (Berea sandstones named Berea 3, 4 and 5) using NMR method, and calculated the NMR-determined relative permeability to water using the Kenyon relationship [34], which was widely used in oilfield NMR logging. The NMR correlation result of Berea 5 sandstone was consistent with formation of hydrate in the centers of pores, rather than with hydrate coating the grains. Kneafsey et al. [35] used fine F110 silica sand and sand/silt primarily in the 100 to 200 µm grain size, and measured methane gas relative permeability. They observed the spatial distribution of water saturation using CT scanning, and found that the two best-fitting models were based on pore filling type rather than grain coating reported earlier [36,37]. In other words, the hydrate crystal occupied the pore bodies instead of occurring on the surface of the sand grains.

In recent years, numerical studies of the permeability of porous media in the presence of hydrate have been reported, including the permeability reduction models and pore network models. Masuda et al. [38,39] introduced a widely used model with a permeability reduction exponent N to relate the permeability of porous media in the presence of hydrate $k(S_H)$ to the absolute permeability of porous media in the absence of hydrate k_0 and hydrate saturation S_H : $k(S_H) = k_0(1 - S_H)^N$, in saturated systems (i.e., $S_G = 0$ and $S_w = 1 - S_H$) $-S_H$). The value of N varies from 1 to 25, and there could be drastic reduction in the permeability when the value of N is taken too large. It may be increased when taking into account the hydrate formation in pore throats. However, there is a debate on the actual value of *N* to be used in the modeling studies. Liang et al. [40] developed a three-dimensional cubic pore network model and studied the effect of the hydrate particle formation and its growth habit on the permeability. The results indicated that the pore-size distribution and the equilibrium temperature for pores with different sizes affected the permeability with different hydrate saturations. Dai et al. [41] developed a pore network model to describe the relationship between the water permeability and the hydrate saturation, accounting for hydrate pore-scale growth habit and heterogeneity. Their model permitted modification of the Kozeny-Carman equation, and they established a relationship between the hydrate saturation and the multiplying tortuosity and specific surface ($\tau \cdot s_s$). Sakamoto et al. [42] developed a relationship between the absolute permeability with methane hydrate formation and the original permeability without any hydrate formation, which was similar to the equation proposed by Masuda et al. [38,39] mentioned above. They related the permeability reduction exponent *N* to the irreducible hydrate fraction S_{H1} , when the hydrate saturation S_H was smaller than S_{H1} . They measured the water effective permeability k_w with methane hydrate in three types of sand columns (Toyoura sand, No. 7 silica sand and No. 8 silica sand). The parameters in the numerical models needed to be obtained from the experimental data, which were quite different due to various experimental conditions, including porous media (e.g., sands, silt, clay, glass beads, and various other field or artificial sediments), gas (methane, CO₂, nitrogen, etc.) used to form hydrate and measure permeability, aqueous (water and other liquid), hydrate saturations and accumulation patterns and so on.

Previous studies reported experiments on the permeability of CO_2 and methane hydrate-bearing sediments. Delli et al. [43] measured the water relative permeability of the Ottawa 20/30 sand samples containing CO_2 hydrate, and the sand grains had a particle size distribution between 600 and 850 µm (mean size of 720 µm). They developed a hybrid model based on the weighted combination of grain coating and the pore filling models, which was used to fit the measured experimental data. Kumar et al. [44] developed an experimental setup to measure the permeability of glass beads with diameter ranging 0.0889–0.14986 mm in the presence of CO_2 hydrate using the stable CO_2 gas flow injection method. The absolute permeability of the porous media in the absence of hydrate was measured to be 66.9 Darcies. They employed the Masuda's model to compare with the experimental data. The permeability

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