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Effect of gas hydrate formation and decomposition on flow properties of fine-grained quartz sand sediments using X-ray CT based pore network model simulation

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ABSTRACT

Experiments are performed to study the closed-loop effect of gas hydrate formation and decomposition on the flow properties of a fine-grained quartz sand specimen. The high resolution X-ray CT images of the test specimen at different experimental stages are acquired. In order to elucidate the changes in pore structure of the test specimen, topologically representative pore networks are established. The evolution of the flow properties during gas hydrate formation and decomposition is further evaluated. The results show that, gas hydrates occupancy in pore space exhibit different modes; they grow mainly as the grain-cementing mode except some intermediate stages, where pore-filling or load-bearing hydrates are observed. It is also found that the formation and decomposition of gas hydrates can cause the pore structure and flow properties changed. Increase of gas hydrate saturation results in a sharp decline in water relative permeability, larger irreducible water saturation and smaller gas-water percolation zone, while gas relative permeability does not exhibit obvious changing law. The decomposition of gas hydrates will exert a greater influence on the flow properties described above than gas hydrate formation does. The increase in frequency percentage of 10–20 μm pores within the fine-grained test specimen after experiments might be caused by gas hydrate decomposition induced damage of pore structure.

1. Introduction

Natural gas hydrates are ice-like crystalline solids composed of hydrogen-bonded water cages and guest hydrocarbon gas molecules. Due to their stability under low temperature and high pressure conditions, the naturally occurred gas hydrates are primarily distributed in the sediments of slopes at marine continental margins and beneath the arctic permafrost [42,43]. Their potential as one of the most promising alternative energy resources, hypothetical impact on global climate, threat to transportation of hydrocarbons in pipelines or wellbores and their considerable capacity for gas storage, have drawn significant attention of extensive research [3,32]. The amount of methane gas estimated in oceanic hydrate-bearing deposits is nearly two orders of magnitude greater than that in arctic permafrost regions. However, efficient utilization of oceanic gas hydrates strongly depends on technical and environmental feasibility of long-term gas production from naturally occurred hydrate-bearing deposits, and plenty of challenges and open issues are still imperative to resolve [40,19,52].

Makogon [29,30] identified three major methods to develop hydrate-bearing deposits: depressurization, thermal stimulation and injection of inhibitors. Depressurization has been the most promising method in terms of potential flow rates from previous gas production tests [26,6,7]. Four drilling expeditions for gas hydrates have ever been performed at the northern South China Sea (SCS), and several huge hydrate-bearing deposits were discovered. Large quantities of natural hydrate-bearing samples have also been recovered. On May of 2017, the first offshore gas production test on hydrate-bearing deposits in Shenhu area, SCS was performed using an improved depressurization method. Satisfactory achievements were reported in terms of the highest cumulative gas production, longest duration time and negligible

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environment effect. Preliminary studies [48,50] showed that the naturally occurred hydrate-bearing deposits in Shenhu area, SCS were dominated by unconsolidated clayey sediments with extremely fine grains and low permeability. Gas hydrates with saturation ranging from 17% to 64% are widely distributed in the fine-grained clayey sediments, which is inconsistent with the knowledge that highly-saturated gas hydrates usually exist in coarse-grained sand deposits and are more desirable for long term gas production [16,46,49]. Due to the unique sedimentary environments, accumulation mechanisms, occurrence patterns and spatial distribution of gas hydrates in Shenhu area, a completely different gas production performance is achieved. The fluid flow behavior, fluid distribution, and productivity at naturally occurred hydrate reservoirs are sensitive to flow properties of hydrate-bearing sediments, which are primarily affected by sediment microstructure, gas hydrate morphologies and saturation in pore space. In order to accurately predict gas production potential and design reasonable development scheme for hydrate reservoirs, it is essential to study the closed-loop effect of hydrate formation and decomposition on flow properties of fine-grained hydrate-bearing sediments.

The characteristics representing fluid flow in pore geometries are composed of intrinsic permeability, relative permeability and capillary pressure. The intrinsic permeability is essentially related to sediment microstructure, while relative permeability stands for a capability of certain fluid flow through the porous medium when multiphase fluids coexist in pore space. The capillary pressure is defined as pressure drop at the phase interface between non-wetting and wetting phase fluids, which is also critical for better understanding the sediment microstructure and multiphase flow behavior. Nowadays, the widely accepted methods to estimate the flow properties in presence of gas hydrates include laboratory measurement, various empirical models and pore network simulation. Direct measurement of flow properties in hydrate-bearing sediments has long been a challenge because of the following difficulties [37,36,20,9]: 1) recovering natural hydrate-bearing samples with pressure coring devices, 2) requiring long induction time for hydrate formation and poor control of gas hydrate saturation due to transient hydrate dissociation and formation during water and gas flow, 3) covering the entire range of gas hydrate saturation using a particular hydrate formation method, and 4) stabilizing gas hydrates under low temperature and high pressure conditions. In absence of reliable experimental data, various empirical models can be used to describe the effect of gas hydrate saturation on flow properties in coarse- or fine-grained hydrate-bearing sediments [23], and the widely used empirical models mainly include parallel capillary model [22], Kozeny-Carman (KG) model [38], Tokyo model [31], van Genuchten [41] and a hybrid model with the combination of grain-coating and pore-filling KG model [8]. Since the majority of empirical models assumed constant pore structures, relevant studies targeted idealized gas hydrate formation or decomposition in simple pore geometries with homogeneous hydrate pore morphologies. However, the hypothesis might induce significant error without considering the effect of hydrate microstructure evolution, capillarity and complicated pore structures.

In recent years, pore network modeling has been rapidly developed from understanding displacement processes [51,45] to providing in-depth digital core analysis with micro-focus CT [10,34]. On the basis of raw CT images, a topologically representative pore network is firstly extracted. Consequently, the relevant displacement and transport equations are computed, and dynamic evolution in permeability and capillary pressure will be analyzed. Due to high computation efficiency, it has been widely applied to the areas of phase exchange, non-Newtonian displacement, non-Darcy flow, reactive transport and CO_2 geological sequestration [5,28]. Jang and Santamarina [18] used pore network models defined with a minimal set of physically meaningful pore-scale parameters to elucidate gas recovery and water production. The effect of fluid expansion, initial hydrate saturation and pore size distribution on recoverable gas and evolution of gas saturation during hydrate depressurization-induced dissociation were further evaluated. Dai and Santamarina [11] employed pore network modeling to determine the capillary pressure curve for hydrate-bearing sediments. The results indicated that gas hydrates in pore space led to higher capillary pressure curves, and the air entry pressure is relatively lower in sediments with patchy distributed hydrate rather than randomly distributed ones, with higher pore size variation and pore connectivity or with lower specimen slenderness along the flow direction. Dai and Seol [12] also evaluated the effect of gas hydrate saturation on hydraulic tortuosity and specific surface area, and a KC-based permeability reduction model was finally proposed. Wang et al Wang et al. [46,47] combined X-ray CT observation and pore network models to compute gas-water relative permeability curve in hydrate-bearing porous media and explored the effect of particle size and wettability on relative permeability. Mahabadi et al. [33] extracted a 3-D pore network with CT images of Mallik hydrate-bearing sediments for pore-scale modeling in order to obtain the proper fitting parameters of capillary pressure and relative permeability curves. Mohammadmoradi and Kantzas [34] introduced a novel three-phase pore morphological simulation approach to simulate hydrate deformities and predict fluid occupancies and permeability of hydrate-bearing geological formations. However, previous studies mainly focused on simple pore geometries or coarse-grained samples restricted to the formation or decomposition of gas hydrates. It is urgent to study the effect of gas hydrate formation and decomposition on the flow properties of fine- grained hydrate-bearing sediments, which may provide new insights into the direction of long-term gas production and utilization in actual fine-grained clayey gas hydrate- bearing deposits.

In this study, the closed-loop effect of gas hydrate formation and decomposition on flow properties of fine-grained quartz sand sediments is studied combining X-ray CT observation and pore network models. This paper is organized as follows: Firstly, experiments for hydrate formation and decomposition in a fine-grained quartz sand specimen are conducted, and X-ray CT images of the test specimen at different experimental stages are acquired. On the basis of raw CT images, topologically representative pore networks are extracted, and the effect of gas hydrate formation and decomposition on flow properties of the test specimen is evaluated. Finally, we summarize the results and present the conclusion of this work.

2. CT experiment for hydrate formation and decomposition

2.1. Experimental setup

The *in situ* X-ray CT experiments for gas hydrate formation and decomposition are performed with a micro/nanometer X-ray CT system, provided by GE Phoenix Corporation. The overall X-ray CT experimental setup consists of a pressure control system, a semi-conductive temperature control system, an X-ray CT scanner and a high-pressure reaction vessel, as shown in Fig. 1. The reaction vessel manufactured with peculiar aluminum alloy materials is 10 mm in diameter. The thickness of the vessel is thin enough to guarantee the X-rays penetrating through the inner structure of test specimen, and endure high pressure for hydrate formation. Besides, there exists a vacuum thermal insulating layer to maintain a fixed temperature. The semi-conductive temperature control system, which is primarily made up of a thermo regulator, a semi-conductive electronic refrigerator, several heat sinks, an electric fan and one Pt100 temperature sensor within ± 0.1 °C, is placed at the bottom of test specimen to monitor the temperature during experiments. The pressure control system denotes to a gas cylinder, a gas compressor and a pressure transducer interconnected with the vessel for pressure data collection. The pressure is actively regulated within ± 0.1 MPa. High resolution CT images are acquired by the X-ray CT scanner which mainly composes of a micro/nanometer alternate X-ray source with monochromatic beam energy of 240 kV or 180 kV, a rotating scanning platform and a highly sensitive flat panel detector. The CT scanner can offer a visualized 3-D distribution of sediment