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# Experimental study on sediment deformation during methane hydrate decomposition in sandy and silty clay sediments with a novel experimental apparatus



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#### HIGHLIGHTS

• A novel 3-D high-pressure reactor with a quick-opening component is adopted.

• CH<sub>4</sub> hydrate decomposition in sandy and natural silty clay sediment is studied.

• Pressure, temperature and gas production behaviors in both sediments are studied.

• Hydrate in silty clay sediment shows a dynamic decomposition condition.

• A radial shrinkage effect of hydrate decomposition is observed in both sediments.

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#### ABSTRACT

Extraction of methane from hydrate-bearing sediment (HBS) has aroused increasing interest around the world. However, the decomposition of gas hydrate may cause significant sediment deformation, which is one of the main obstructions for the hydrate exploitation. To investigate the decomposition behaviors of methane hydrate in different sediments and study the sediment deformation during hydrate decomposition, two contrast experiments were carried out in a novel three-dimensional (3-D) high-pressure reactor with a quick-opening component. In this study, synthetic sandy sediment and natural silty clay sediment sampled from the Shenhu Area, South China Sea were used as the HBS. For hydrate in sandy sediment, the pressure-temperature (P-T) relationship is consistent with that of bulk hydrate. However, in silty clay sediment, the hydrate decomposition conditions shift into a higher pressure region than that in sandy sediment, which is mainly due to the small pore particle size and the presence of salinity. In addition, hydrate decomposition conditions in silty clay sediment also vary at different positions in the reactor because of the effects of salinity, organics, minerals and the uneven distribution of the pores and pore particles. Hence, it is considered that there is a variable equilibrium for the methane hydrate deposits in silty clay sediment. Further, Radial Shrinkage Effect of Hydrate Decomposition (RASHEHD) was observed in both experiments. It is possibly a combined consequence of gas seepage and the cementation effect of hydrate.

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#### 1. Introduction

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Gas hydrates are solid crystalline compounds in which gas molecules as guests occupy the lattice structure formed by  $H_2O$  [1]. The most common type of gas hydrates occurring naturally mainly contains CH<sub>4</sub>, usually called natural gas hydrate (NGH). NGH is considered to be one of the most important potential



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energy sources in the 21st century. As estimated, the energy reserve of the NGH is twice as great as the combined traditional fossil fuels on land, including petroleum, coal and natural gas [2]. To keep the stable presence of hydrate, relatively low temperature (T) and high pressure (P) are the two necessary conditions. Thus NGH deposits mostly occur in the permafrost and the marine sediments, and the reserve in the ocean is far larger than the total reserve in the permafrost [3,4].

Four practical methods for the recovery of natural gas from NGH reservoirs are: (1) thermal stimulation method [5–7], (2) depressurization method [8–11], (3) chemical inhibitors injection [12], and (4) CO<sub>2</sub> replacement method [13,14]. To obtain better gas production rate and energy efficiency, Li et al. [15] and Song et al. [16], carried out a series of experiments with combined recovery methods, such as huff and puff in conjunction with depressurization, huff and puff with no soaking in sandy sediments and depressurization with heat stimulation. Furthermore, some experimental and numerical models were developed to study the production behaviors of different well spacing, which includes single horizontal well, dual horizontal wells and five-spot well system [17–20]. Through these studies, the four gas production methods were verified and optimized in different scales.

It is believed that NGH reservoirs commonly occur in unconsolidated sediments without adequate shear strength. The decomposition of hydrate may cause significant sediment deformation and reduce the shear strength of the sediments [21]. In fact, some efforts have been made to understand the deformation mechanism and the mechanical properties of the hydrate bearing sediments (HBS). Sultan et al. [22,23] found that the melting of NGH would generate excess pore pressure and reduce the soil resistance. By hydrate decomposition experiments with depressurization method, Kono et al. [24] derived a kinetic decomposition rate equation and found that the decomposition rate was controlled by the sediments properties. In the work of Haligva et al. [25], variable-volume bed is correlated to the gas production behaviors. Jung et al. [26] carried out several methane hydrate decomposition experiments in sediments with different fractions of fine particles. They found a particle migration with fluid flow and a fracture structure formed in sandy sediment, and they defined the critical fines fraction to describe the effects of the fine particles. In the work of Seo et al. [27], an experimental model was established in sediments of small pore gels, in which a phase equilibrium shift of hydrate towards a higher pressure region was confirmed. Recently, to study the mechanical properties of HBS, some mechanical tests have been carried out in triaxial [28,29] and centrifuge apparatuses [30,31] using Tetrahydrofuran or CO<sub>2</sub> hydrate. Through the study of the vertical and shear deformation of HBS, several strain-stress constitutive models have been derived [32].

Till now, few have studied the differences between the hydrate decomposition characteristics in synthetic sandy sediment and natural silty clay sediment. Moreover, little knowledge has been known about how hydrate decomposition acts on the sediment deformation. Thus it is significant to observe the sediment deformation directly, which will provide a firm foundation for further analysis.

This work is aimed to investigate the decomposition behaviors of methane hydrate in different sediments and analyze how hydrate decomposition acts on the sediment deformation by observation. Synthetic sandy sediment and natural silty clay sediment were used as the HBS in this work. The silty clay supplied by Guangzhou Marine Geological Survey (GMGS) was sampled at a depth of 1400 m below the sea level, from the Shenhu Area, South China Sea. In this study, an innovative 3-D high pressure reactor with a quick-opening component was used to reproduce the hydrate formation conditions in the survey area.

#### 2. Experimental section

#### 2.1. Experimental apparatus

The experimental apparatus which was used in this study is schematically shown in Fig. 1. The experimental apparatus consists of a cubic high-pressure reactor equipped with a quick-opening component, a temperature-controlling water bath, an output unit, a back-pressure regulator, a gas-liquid-solid three-phase separation system, a data acquisition system, and some measurement units. The cubic three-dimensional (3-D) high-pressure reactor is made of 316 stainless steel and can hold a pressure up to 30 MPa. The edge length of the cubic reactor is 90 mm, and the effective volume of it is 729 ml. The quick-opening component of the reactor involves a pair of stainless steel clamps connecting the top cover with the reactor and a rubber O-ring set on the top cover to seal the reactor. At the bottom of the reactor, there are 27 Pt100 thermocouples with a measuring range from 223.15 K to 473.15 K and an accuracy of ±0.1 K. They are evenly distributed at nine measuring points and divided into three layers, namely, top (A), middle (B), and bottom (C). Corresponding to the positions of the 9 measuring points, 9 wellheads for vertical wells are set on the top cover. The layout of the temperature measuring points and wellheads is shown in Fig. 2. In this study, wellhead 5 was used as the production well and other wellheads were sealed. The inlet is set at the center of the reactor's bottom. The reactor is placed in a temperature-controlling water bath to keep a constant ambient temperature. Two pressure transducers (TRAFAG NAT 8251.84.2517 type, measuring range of 0-25 MPa, ±0.02 MPa) are employed to measure the inlet and outlet pressures. The output unit is connected with a desander to separate the solids from the produced gas and water, the working pressure of which ranges from 0 to 40 MPa. A back-pressure regulator supplied by the TES-COM Co. is connected to the outlet of the desander to control the production pressure. The controlling range of it is 0-30 MPa, ±0.02 MPa. The mass of the liquid produced from the reactor is measured by an electronic balance (Santorius BS 2202S, 0-2200 g, ±0.01 g). A gas flow meter supplied by the Seven Star Co. (D07-11CM type, the measuring range of 0-10 L/min,  $\pm 2\%$ ) is employed to record the gas production rate and the cumulative gas production. The thermocouples, pressure transducers and the gas flow meter were calibrated using a mercury thermometer with a tolerance of ±0.01 K, a pressure gauge with an error of ±0.05%, and a wet gas meter with an accuracy of ±10 ml/min, respectively. Then the temperature, pressure, volume of the cumulative gas production, gas production rate, and mass of the liquid are recorded by the data acquisition system. Furthermore, deionized water is used in this work. The methane with a purity of 99.999% is supplied by Fushan Hua Te Gas Co.

#### 2.2. Materials

Silica sand and silty clay were used as the sediments in this study. The silica sands used in this study were obtained from the Bandao Silica Sands Co. The silty clay supplied by Guangzhou Marine Geological Survey (GMGS) was sampled at a depth of 1400 m below the sea level from the Shenhu Area, South China Sea. Some measurements of the sediments' properties were made here. Firstly, the two kinds of sediments were dried thoroughly in a drying oven at 393.15 K for more than 48 h. Then the density of the dry samples was measured by the true density meter (VPY-30, Quantachrome). Subsequently, the specific surface area and the particle pore volume were measured based on the multi-point BET analysis by ASIQMO002, Quantachrome. Finally, the particle diameter distributions were determined using the Mastersizer

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