



# Decomposition conditions of methane hydrate in marine sediments from South China Sea



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## ARTICLE INFO

### Article history:

Received 1 August 2015

Received in revised form

26 November 2015

Accepted 2 December 2015

Available online 24 December 2015

### Keywords:

Methane hydrate

Marine sediments

Equilibrium decomposition condition

## ABSTRACT

Decomposition conditions of methane hydrate in marine sediments from South China Sea were measured using multi-step decomposition method. Four different samples of the marine sediments were used in the experiments. The pore distribution, the surface area, particle size and the surface texture were measured and observed. The experimental results indicated that the final decomposition temperatures are shifted lower than those for bulk hydrates at the same pressure for different marine sediments and different water saturations. Temperature shifts are more negative for smaller initial water saturation. The surface textures and pore of the sediments both affect the equilibrium condition of methane hydrate. Using the Clausius-Clapeyron equation, the enthalpy of hydrate dissociation in marine sediments was calculated. It was found that the enthalpy of hydrate dissociation in marine sediments with lower water saturation is higher than that at the bulk state.

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## 1. Introduction

Gas hydrate is vastly distributed throughout both marine and permafrost areas, and represents a potential huge resource [1–3]. Gas hydrates are generally distributed in marine sediments along continental margins. The pore distributions of the marine sediments will raise the hydrate formation and decomposition pressure compared with the bulk hydrate at a same temperature. Besides, the different mineral composition of the particles and the water saturation will also affect the equilibrium hydrate decomposition conditions [4]. Phase equilibrium studies of natural gas hydrate systems in marine sediments are of significance for assessing the reserves of the natural gas hydrate, finding the location of the bottom of the hydrate stability zone, and providing the basic data for determining the exploitation condition [5].

Handa and Stupin [6] first measured the equilibrium dissociation conditions of methane hydrate and propane hydrate in porous media. After that, many researchers performed the experimental studies to investigate the effects of the porous media on the hydrate equilibrium decomposition conditions [5,7–10]. It was found that the hydrate forms and decomposes at lower temperatures or higher

pressures in mesoporous media compared with that under bulk condition attributing to capillary-pressure-induced depression of the water activity.

Although hydrate formation conditions in artificial porous media are relatively well known, the studies on the hydrate decomposition conditions in marine sediments are still limited. The physical and chemical properties of the marine sediments are usually very complicated, which may consist of sand, clay, organic matter, carbonates, silicates and other mineral components [11]. Therefore, the experimental data obtained in artificial porous media is difficult to be directly applied to the prediction of the decomposition conditions in marine sediments. In order to investigate the effect of the complicated composition of the sediments on the hydrate decomposition behaviors, several studies have been taken. Uchida et al. [11] measured the decomposition conditions of methane hydrate in sediments. The silica sand, sandstone, and clays (kaoline and bentonite) were used in the experiments. They concluded that the decomposition conditions are affected mainly by the pore sizes. The results indicated that hydrates form not only between the particles but also in the interlayer of the bentonite. Saw et al. [12] experimentally investigated into the effect of bentonite clay on methane hydrate formation and dissociation behaviors in synthetic seawater. It was found that hydrate equilibrium decomposition conditions are effected by the concentration of bentonite clay in synthetic seawater. Yakushev [13] measured the

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hydrate formation conditions in the clay medium and found the importance of clay on hydrate formation. They found that low water contents inhibit hydrate formation in spite of having good permeability in the unconsolidated clay powder. The hydrate accumulation in the same clay sample increases with the increase of the water content. Clennell et al. [14] found that the particle size of the marine sediments also affect the hydrate equilibrium decomposition through the experiments. Lu and Matsumoto [15] investigated the stability conditions of methane hydrate in a nanofossil-rich claystone column. They found that the equilibrium decomposition temperature of methane hydrate at a given pressure in the test sediment column is shifted to the lower temperature field by 0.4 °C and 1.5 °C compared to those in sea water and pure water, respectively.

In 2007, it was announced that the natural gas hydrate samples was firstly recovered at the water depth 1500 m and 200 m below seafloor level in the Shenhu area of South of China Sea [16]. The hydrate layer in the Shenhu Area was found to be in the pressure range of 12.7–14.6 MPa, the temperature range of 8.6–16.2 °C, and the hydrate saturation range of 1.0–47.3% with an average value of around 22% [16]. Most gas hydrate samples so far collected in this area were bacterial methane gas hydrate type I. Sun et al. [17] found that the equilibrium temperature of methane hydrate in the South China Sea sediment sample with in-situ pore water is decreased, and it is controlled by the pore water. However, it is still lack of the studies of the effect of the complicated physical properties on the hydrate equilibrium conditions in marine sediments [18], particularly of the marine sediments from the Shenhu area, South Sea of China.

To provide the experimental data for using in future hydrate exploitation from marine sediments, and to gain a better understanding of the hydrate decomposition properties in marine sediments, we have performed experiments and investigated the properties of methane hydrates in four samples of marine sediments from the South Sea of China. The equilibrium hydrate decomposition conditions were measured using the multi-step heating dissociation method. The samples have different pore distributions, particle diameters and specific area, which should help to understand the effect of the texture of the marine sediments on the hydrate decomposition. The effect of the water saturation on the decomposition conditions was also investigated with the various water contents in sample.

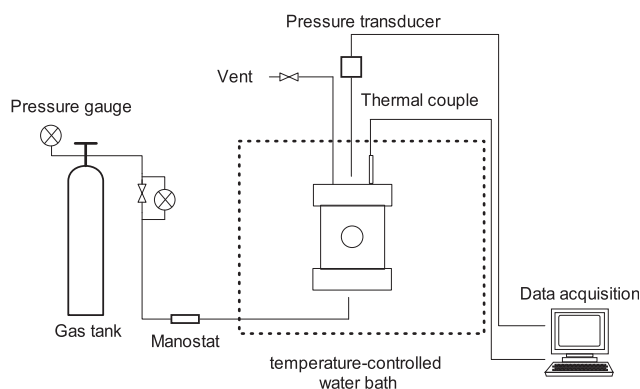


Fig. 1. Schematic of the experimental apparatus.

## 2. Experimental setup, materials and procedure

### 2.1. Apparatus

The experimental apparatus, shown in Fig. 1, consists of the high-pressure hydrate crystallizer (CR), the gas/liquid supply system, the temperature-controlled water bath, the pressure control system and the data acquisition system. The effective maximum volume of the cell is 416 cm<sup>3</sup>. The pressure in the cell of the crystallizer is measured using a MBS3000 absolute pressure transducer with the accuracy of  $\pm 0.02$  MPa for the complete operating range of 0–25 MPa. A Pt1000 thermoprobe (JM6081) measures cell temperature within a precision of  $\pm 0.05$  K. The crystallizer temperature is controlled by a temperature-controlled water bath within a range of 263.15–303.15 K and can be kept stable to with  $\pm 0.1$  K. The signals of the pressure and the temperature are continuously monitored and recorded by a data acquisition system coupled with a computer. The methane with the purity of 99.9% was obtained from Fushan Hua Te Gas Co.

### 2.2. Materials

Four kinds of marine sediments were used in the study. The marine sediments supplied by Guangzhou Geological Survey were from the Shenhu Area in the South China Sea. The density of the dry samples was measured by the true density meter (VPY-30, Quantachrome). The particle diameter distributions of the dry samples were measured by the Mastersizer 2000 particle size analyzer (Malvern Instruments, Ltd.). The measured particle diameter distributions are shown in Fig. 2 and the physical properties of each sample are listed in Table 1. As a representative, the diameters of Sample 1 ranged from less than 0.399 to 178.25  $\mu\text{m}$  with an average diameter of about 7.898  $\mu\text{m}$ . Specific surface area and pore size analyzer (ASIQM0002-2, Quantachrome) were used to measure the distributions of pore volume and the specific surface area of the samples, respectively. Pore volume was determined based on the BET analysis. The measured pore diameter distributions are shown in Fig. 3. The pore volume and the specific area of each sample are also listed in Table 1. The micrographs of the four samples were tested via the scanning electron microscopy (Hitachi S4800 FESEM). Fig. 4 gives the electron micrograph of Sample 3. It can be seen that the marine sediments have complicated configurations and physical properties. Diatoms were also observed among the sediment particles.

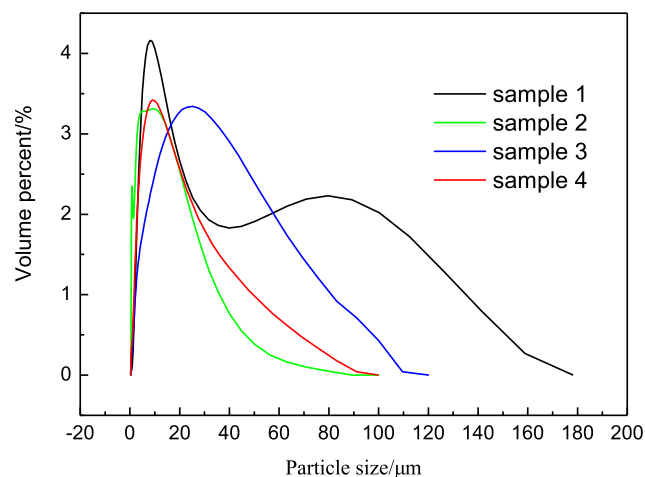


Fig. 2. Particle size distributions of different marine sediments.

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