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PORE-SCALE INVESTIGATION OF GAS HYDRATE DISTRIBUTION CHARACTERISTICS IN POROUS SEDIMENTS WITH MICRO X-RAY CT

Xuan Kou [a,b,c,d,e] ,Yi Wang [a,b,c,d] , Xiao-Sen Li [a,b,c,d,*]

a. *Guangzhou Institute of Energy Conversion, Chinese Academy of Sciences, Guangzhou 510640, P. R. China.,*
b. *CAS Key Laboratory of Gas Hydrate, Guangzhou 510640, P. R. China.,*
c. *Guangdong Provincial Key Laboratory of New and Renewable Energy Research and Development, Guangzhou 510640, P. R. China.,*
d. *Guangzhou Center for Gas Hydrate Research, Chinese Academy of Sciences, Guangzhou 510640, P. R. China.,*
e. *University of Chinese Academy of Sciences, Beijing 100083, P. R. China.*

ABSTRACT

Natural gas hydrate has been regarded as an alternative energy resource and has attracted much attention in recent years. According to laboratory investigations and field test researches, pore-scale distribution habit of gas hydrate in the hydrate bearing sediments plays a critical role in gas hydrate exploration and exploitation. In this work, the micro X-ray computed tomography was applied to investigate the pore-scale hydrate distribution habit during hydrate formation and dissociation. The gas hydrate was formed and dissociated in-situ and a nondestructive detection was performed to observe the internal characteristics of the hydrate sediment. The experimental results indicate that the hydrate distribution habit evolves from grain-coating to pore-filling during hydrate formation. During hydrate dissociation, thermal stimulation can promote homogeneous hydrate redistribution, and uniform distribution of glass beads is promoted by the depressurization.

Keywords: Gas hydrate, micro X-ray CT, distribution habits, formation, dissociation

1. INTRODUCTION

Natural gas hydrate, buried under the sea and the permafrost, has been regarded as an alternative energy resource and has attracted much attention in recent years. As is well known, a large amount of methane gas is stored in the gas hydrate sediments [1]. In order to exploit the gas hydrate which has great resource potentiality, exploration methods such as seismic waves, and thermal conductivity measurements have been implemented in gas hydrate exploration [2]. In addition, main exploitation methods such as depressurization [3], thermal stimulation [4], the combination of the two methods [5] and solid fluidization have been proposed to produce gas from hydrate bearing sediments.

Pore-scale distribution of gas hydrate in the sediments plays a critical role in gas hydrate exploration and exploitation. On the one hand, the spatial distribution of hydrate phase in the sediments has effect on physical properties of hydrate bearing sediments [6]. On the other hand, the distribution habit of hydrate phase in porous sediments will determine the relationship between permeability and hydrate saturation in the capillary tube model [7]. Therefore, it is important to understand the pore-scale distribution habits of gas hydrates during hydrate formation and

[*] Corresponding author. Tel: +86-20-87057037; fax: +86-20-87034664.
E-mail address: lixs@ms.giec.ac.cn. (X.-S. Li)

dissociation, for either experimental research or field test of hydrate exploitation.

In recent years, X-ray computed tomography (X-ray CT) has been an effective way to study the distribution characteristics of hydrate in the porous media. Kerkar et al. [8] used synchrotron X-ray computed tomography (CMT) to visualize methane hydrate growth for the first time, and they found that the pore-filling hydrate formed from dissolved methane gas. In addition to typical guest molecules, the high atomic number gas molecule, xenon, was also served as a proxy in X-ray CT detection of in-situ synthesized hydrate [9]. Chaouachi et al. [10] studied the growth and dissociation of xenon hydrate in three scenarios with different amount of gas dissolved in water. They found that the hydrates with different initial growth morphologies appeared to perform similar arrangements in the end of hydrate formation. It is generally accepted that the hydrate saturation has an essential effect on hydrate distribution habit which evolves from grain-attaching to pore-filing or even grain-cementing as hydrate saturation increases. A major current focus in the hydrate distribution habit is the time-resolved evolution of hydrate phase during hydrate formation and dissociation. However, few researchers have addressed the effect of different dissociation methods on hydrate distribution characteristics.

In this work, gas hydrate is formed and dissociated in a cylinder reactor with an effective volume of 3.925 mL to investigate the hydrate distribution habits during hydrate formation and dissociation. The time-resolved distribution habits of hydrate phase in the pore scale are investigated by the micro X-ray CT. Meanwhile, xenon gas that has similar properties with methane gas is used as the guest molecule of hydrate to enhance the image contrast.

2. EXPERIMENTAL SECTION

2.1 Experimental apparatus

The schematic of the experimental systems applied in this work is shown in Fig. 1. The experimental system consists of three primary units: in-situ hydrate synthesis unit, X-ray CT inspection unit and computer unit.

In-situ hydrate synthesis unit comprises four main parts: (1) a cylinder reactor with an effective volume of 3.925 mL; (2) a gas cylinder to supply xenon gas for hydrate formation; (3) a temperature controlling system including a water-cooling box, an insulation jacket and a heat insulation chamber; (4) pressure and temperature transducers to acquire the pressure and temperature of the reactor. The X-ray CT inspection unit mainly comprises three parts: (1) two X-ray tubes (micro-focus and nano-focus X-ray tubes) to emit X-ray beams; (2) an X-ray flat panel detector to receive X-ray beams; (3) a sample manipulator with a rotating and moving stage.

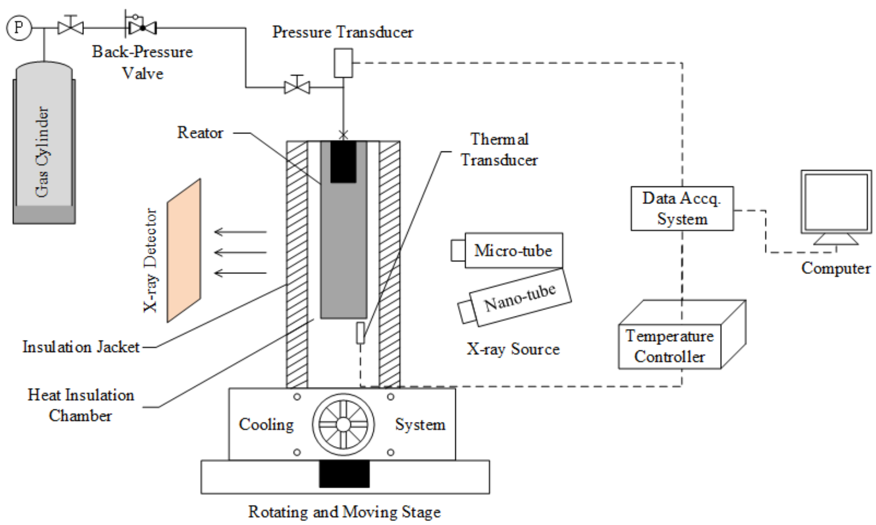

Fig 1 Schematic of the experimental systems consisting of three primary units: in-situ hydrate synthesis unit, X-ray CT inspection unit and computer unit.

2.2 Experimental procedure

2.2.1 Sample preparation

Before hydrate formation, deionized water and glass beads (GB) with diameter of 0.350~0.500 mm were firstly compacted in the high pressure vessel. Thirdly, xenon gas was pumped into the vessel three times to expel residual air. Afterwards, the micro X-ray CT scan was performed to acquire the initial condition of the sample in the reactor. Then the temperature was set to a target value of 276.15 K and kept constant by the temperature controlling system. After three days of hydrate formation, the experimental investigations of hydrate dissociation by thermal stimulation and depressurization were carried out. The hydrate sediment was heated to 2.5 ^{o}C, 5.5 ^{o}C, and 8.5 ^{o}C, respectively. When pressures were stable in each temperature condition, the X-ray CT scans were performed to record the hydrate distribution characteristics. Afterwards, the hydrate sediment was depressurized twice to completely dissociate gas hydrate in the reactor, and each stage was recorded by the micro X-ray CT scan to investigate the hydrate dissociation characteristics.

2.2.2 X-ray CT scan

The X-ray CT equipment used in this work is Phoenix Vtomesx s240/180. The X-ray tube emits X-ray beams that penetrate the sample then hit the detector. Each scan was terminated by 1200 slices of the sample which are acquired in 1 h with the beam energy of 110 keV /100 mA. After each scan, 2-D images were collected by the computer unit, then reconstructed into a 3-D volume and processed by VG studio and ImageJ to show pore-scale hydrate phase distribution habits in the sediments.

3. RESULTS AND DISCUSSION

3.1 Nucleation and growth

Figure 2 shows the time-varying distribution of xenon hydrates in the porous media. As shown in Fig. 2, a large hydrate chunk is accumulated on the top of the reactor because 90% volume of the reactor was filled with porous media. It can be seen from image D that the hydrate crystals tends to nucleate on the surface or the contact of each GB before growing into pores. Thus, during the early stage of hydrate formation, the hydrate distribution habit follows grain-coating or grain-cementing. Meanwhile, further increase in thickness of the hydrate film have transformed the hydrate distribution habit into pore-filling. Therefore, it can be inferred from the time-varying development of hydrate formation that the hydrate distribution habit during the formation process evolves from grain-coating or grain-cementing to pore-filling. In addition, since hydrate formation is an exothermic reaction, the gas hydrates are accumulated around the reactor of relatively high efficiencies of heat transfer. Furthermore, image D in Fig. 2 also exhibits dissociation of the thin hydrate film and accumulation of large hydrate crystals, this phenomenon has been interpreted as the Ostwald ripening.

3.2 Dissociation characteristics

The hydrate sample was dissociated with different methods after hydrate formation. Firstly, free gas in the sediments was released before rising temperature to 2.5 ^{o}C, 5.5 ^{o}C and 8.5 ^{o}C, respectively. After pressure in the reactor remained stable under each temperature condition, the X-ray CT was performed to investigate the distribution characteristics of gas hydrate. Figure 3 illustrates the hydrate saturation versus depth which is from each slice to the top of the reactor. With the increase of temperature, the hydrate chunk gradually dissociates and hydrate crystals re-form in other pore spaces, thus inducing the homogeneous redistributions of gas hydrates. In addition, the hydrates tends to re-form in the bottom of the reactor. This is because that

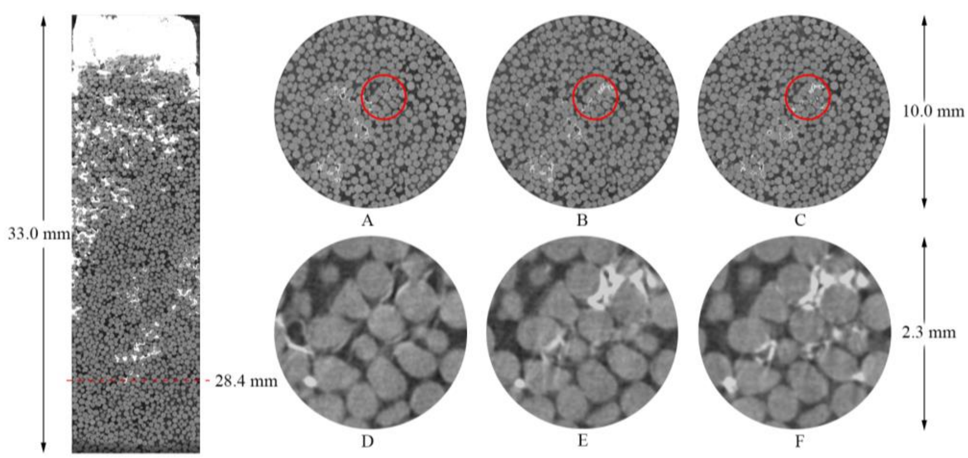

Fig 2 Time-varying distribution of xenon hydrates (bright white) in the porous media of glass beads (light gray) and water (dark gray). An axial CT image and three transverse planes at depth of 28.4 mm; A, B, and C depict the hydrate distribution habits of three hydrate formation stages and D, E, F are the magnifications of red circle in A, B, and C, respectively.

the gas released from the hydrate chunk dissociation migrates from the top to bottom where the P-T conditions are still above the hydrate equilibrium condition. As shown in Figure 3, the hydrate sediment was depressurized twice to dissociate the gas hydrate after thermal stimulation. It is noteworthy that the hydrate chunk has been completely dissociated, and a large amount of secondary hydrate is re-formed in the depth from 10 mm to 20 mm. Furthermore, the depressurization process promotes gas and water fluid to flow from the bottom to top of the reactor, and the re-formed grain-coating hydrate bridges and supports each GB. Thus, the uniform and homogeneous distribution of GB is observed during hydrate dissociation by depressurization.

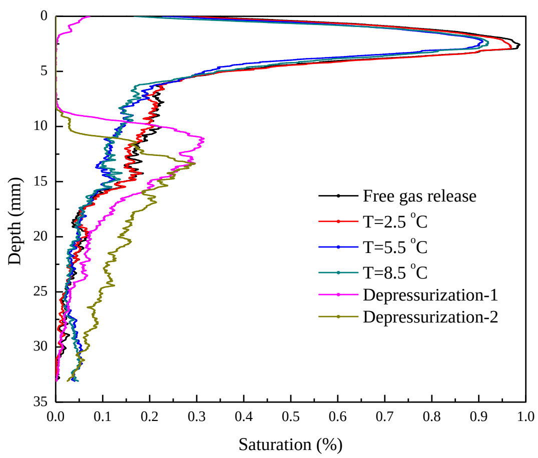

Fig 3 Hydrate saturation versus depth during hydrate dissociation via thermal stimulation and depressurization.

4. CONCLUSIONS

In this work, the micro X-ray computed tomography was applied to investigate the pore-scale hydrate distribution habits during hydrate formation and dissociation. The gas hydrate was formed and dissociated in-situ and a nondestructive detection was performed to observe the internal characteristics of the hydrate sediment. The experimental results indicate that the hydrate distribution habit evolves from grain-coating to pore-filling during hydrate formation. During hydrate dissociation, thermal stimulation can promote homogeneous hydrate redistribution. Besides, the uniform distribution of glass beads is promoted by the depressurization.

ACKNOWLEDGEMENT

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