Holistic Recognition of Methane Hydrate in Marine Sediments by X-Ray CT

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ABSTRACT

Methane hydrate is important energy source in the future. The recognition of methane hydrate in marine sediments is a tough task all the time. We employed a mircofocus X-ray CT to investigate the methane hydrate in South China Sea sediments. The CT images indicate that the various components are difficult to be distinguished because of the limitation of CT resolution, but the change of gray distribution can be used to recognize the appearance of methane hydrate. The formation of hydrates caused the density change of sediment sample, which can be reflected on the CT images. The pixel number change of about 13% may correspond to the hydrate saturation formed in the sediments. The results of this study is of significance to the holistic recognition of methane in marine sediments.

Keywords: Methane hydrate, marine sediment, CT, density

NONMENCLATURE

Abbreviations	
CT MH	Computed tomography Methane hydrate
Symbols	
μm	Micrometer
mm	Millimeter
m	Meter
S	Second
kV	Kilovolt
μA	Microampere

1. INTRODUCTION

Methane hydrate, one kind of crystalline compounds, is thought as an important source of energy in the future decades [1, 2]. It is reported that the distribution locations of MH reservoirs are mainly including the permafrost and marine region [3]. As the advancement of MH research work, China pays more attention on the exploration and exploitation of marine MH reservoirs. Particularly, South China Sea is rich in MH reservoirs in the shallow sediments, about 150 m below the sea floor [4]. In addition, SCS hydrate-bearing sediments are reported to be muddy fine silt clay. However, there are still lots of difficulties for MHs exploitation in the silt clay sediments [5].

Various aspects of MH researches have been conducted in the past decades. The accurate recognition of MH in sediments is of great fundamental significance for MH exploitation. In addition to seismic wave and acoustic wave that are commonly used to detect hydrates in sediments, X-ray CT is another non-contact and nondestructive testing technique by density difference. Recently, the application of X-ray CT on hydrate recognition has drawn more attention. However, it is still difficult to detect the compositions containing hydrates in marine sediments of muddy fine silt clay.

According to exploration results, the grain sizes of clay and silt in marine sediments are less than 4 μ m and 63 μ m, respectively [5]. It should be noted that not all compositions can be recognized by CT apparatus depending on CT maximum resolution and sample properties. This study tried to distinguish the hydrates and describe the hydrate change in marine sediments.

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2. EXPERIMENTAL

In this study, a microfocus X-ray CT (SMX-225CTX-SV, Shimadzu, Japan) experimental system was employed to observe the microstructure and occurrence of methane hydrates in marine sediments [6]. The maximum resolution of 4 μ m/pixel was used in this study. The CT imaging was performed at a source voltage of 120 kV and a current of 50 μ A. The source object distance and source image distance were 46.0 and 1000 m, respectively. A three-dimensional image was acquired after one revolution in 400 s. The images were obtained with a 16-bit gray scale and 2048*2048 pixel resolution.



Fig. 1 Digital photo of marine sediment sample

The procedures of experiments can be described as follows. A 25% moisture of marine sediments were prepared and uniformly packed in CT vessel that is made of polyimide. The marine sediments (see Fig. 1) were obtained from the 122 m depth layer, Lizhi Bay, Shenhu Waters, South China Sea [7]. And then, high-pressure methane gas was charged into the vessel. The formation condition was set at constant 5.5 MPa and 273.65 K. The gas consumption is recorded by a high-precision plunger pump (260D, Teledyne ISCO, USA). The sample was scanned by CT during the whole formation process.

3. RESULTS AND DISCCUSION

The software *Avizo 9.0.1* was used to process and analyze the CT images in this study. A 750*750*400 pixel



Fig. 2 Two-dimentional reconstruction CT image

cuboid, corresponding to 3*3*1.6 mm, inside the sediment sample was selected as the zone of research. Fig. 2 shows the undressed two-dimensional CT image of the research zone. The CT image seemed that the whole sample was a whole, and the pore was invisible. It may be due to the water-absorbing quality of silt clay, causing the gas and water uniformly distributing in the sediment sample. It should be noted that no obvious image change of CT scanning was observed after hydrate formation. In other words, the different compositions of sediment sample cannot be recognized directly.

3.1 Gray distribution

At first, the gray distribution was analyzed, as shown in Fig. 3. It should be noted that the gray distributions of no matter before or after hydrate formation, only one distribution was acquired. In other words, the components and phases in marine sediments cannot be distinguished using our CT apparatus. That was limited by the maximum resolution and affected by the presence of sediments. However, the difference of the gray distribution profiles were obvious before and after hydrate formation. The width and height changes of the distribution profiles represent the sample density and the corresponding amount changes, respectively. And two crossover points at the gray values of 42120 and 43150 appeared in the gray distribution profiles. Based on this, the gray distribution range was divided into three regions. The amount of sample composition of high gray (R3) and low gray (R1) increased, while the amount of sample composition of medium gray (R2) decreased.



Fig. 3 Gray distribution profiles of CT images

The recognition of CT is by the density differences of materials and the gray of CT images inflects the density. In general, the higher the density, the larger the image gray will be. In other words, after the hydrates formed,

the amount of sample composition of high and low densities increased, and the amount of sample composition of medium density decreased. It can be inferred from the results that the density of part sediments increased and part sediments decreased. In fact, the density of solid sediments would not change, however the holistic density of sediments would change. The density of MH is lower than water, so the holistic density of hydrate-bearing sediments decreased as expected. In addition, the sediment has a good absorption of water, so the moisture might affect the density of sediments as well. When some water converted into hydrates, the moisture of part sediments decreased, and the density of the part sediment would increase. The results shown in Fig. 3 displays a comprehensive phenomenon. In R1 zone, the hydrate occurrence decreased the holistic density, i.e. increasing the amount of low density. In R3 zone, the moisture decrease made the density of part sediments increase, inducing the amount increase of high density. In R2 zone, the amount decreased naturally because a lot of them moved to R1 or R3 zones.

3.2 Regional analysis

According to the above division of gray range of CT images, samples in different regions were displayed in different colors. As shown in Fig. 4, the colored images of the same section of vessel before and after hydrate formation represents the amount of different gray values in three regions. In general, the yellow and gray areas increased after the hydrates formed. It should be noted



that the increased yellow area was a little scattered in a certain zone. The main reason is that the MH formation process is believed to be random. In addition, the increased gray area seemed to be close or even connected to the yellow. That confirms that the hydrate formation may cause the density decrease of part sediments and increase of part sediments.

The holistic gray distribution and sectional reconstruction images indicate the formation of MH changed the sediment density [8]. Considering the conversion of watery clay to dry clay and hydrates, it is very reasonable that one material becomes into two, and one is with higher density and the other one is with lower density. Based on this, the X-ray CT can be used to detect the hydrate and change in marine sediments during exploitation process.

3.3 Saturation variation

In order to figure out the detailed change situation, we calculated the saturation variation of three regions by the following two equations.

$$Saturation = \frac{Number of regional pixels}{Number of holistic pixels}$$
(1)

Saturation Variation = Saturation (After MH formation) – Saturation (Before MH formation) (2)



(a) Before formation (b) After formation **Fig. 4** Processed reconstruction images of the same sample section (Yellow: R1, Green: R2, Gray: R3)

Fig. 5 shows the saturation variation of three regions. It is clear that the saturation increase of R1 is a little higher than that of R3, meaning that the gray or density of the holistic sample displays a decreasing result. In addition, the summed increase of R1 and R3 is a bit less than the decrease of R2. In other words, the





conversion of water to hydrate is a density decrease process.

According to the methane gas consumption and hydration number of 5.9, the hydrate saturation of the holistic sample is about 10%. The total increase (R1 + R3) or decrease (R2) of regional saturation is about 13%. The two values are rather close to prove the agreement of CT scanning with gas consumption. And X-ray CT is recommended to be used to detect the hydrate saturation, especially the change during exploitation process in marine sediments.

4. CONCLUSION

A mircofocus X-ray CT of 4 μ m resolution was employed to investigate the methane hydrate in South China Sea sediments with 25% moisture. Direct processing of CT images cannot be used to distinguish the various components because of the limitation of CT resolution, but the change of gray distribution can be used to recognize the appearance of methane hydrate. The formation of hydrates caused the density decrease of part sediments and increase of part sediments. The density change of sediments can be reflected on the CT images in return. The pixel number change of about 13% has a good agreement with the hydrate saturation calculated by gas consumption. The results of this study is of significance to the holistic recognition of methane in marine sediments using X-ray CT.

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