Article

Microcosmic Characteristics of Hydrate Formation and Decomposition in the Different Particle Size Sediments Captured by Cryo-SEM

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Abstract: Sand production and sand control studies in non-diagenetic reservoirs are the weak point in the conventional petroleum industry. However, natural gas hydrate (NGH) mainly exists in non-diagenetic strata, and sand production occurs during exploitation, which restricts the safe and sustainable production of NGH. To study the microcosmic characteristics of sand production, the hydrate decomposition behaviours in the sediments were captured by the Cryo-SEM method. The micromorphology of different particle sizes of sand samples containing NGH (the sand median diameter d50 is 150, 87, 55, 38, 24, and 13 μm) and the microcosmic processes of NGH decomposition were observed. Then, the microcosmic characteristics of sand production, during the decomposition process, were analysed. (1) The gas hydrate decomposition increases pore space and reduces reservoir strength; the expansion action of the decomposition of water and gas, the softening action of the decomposition of water, and the compression action by overlying stress (crustal stress) promoted sand production, deformation, and subsidence of the NGH reservoirs in the mining process. (2) The decomposition of NGH has a more significant impact on sediments with smaller particle sizes. (3) The particle size of NGH may be larger than the particle size of the mud in the reservoirs, and acting as “gravel” plays a particular role in sand control. Therefore, the particle size of NGH cannot be ignored in the design of sand control. (4) It has been revealed, and verified, that sputtering (splashing) is a unique process of sand production caused by NGH decomposition. In other words, the rapid expansion of the volume of the decomposed gas and water from NGH leads to the eruption and sputtering of hydrate particles, providing the driving force for sand migration, which is a different process of sand production than in conventional oil and gas.

Keywords: gas hydrates; decomposition; sediments; Cryo-SEM

1. Introduction

Natural gas hydrates are crystalline solids, composed of water and gas, widely distributed in deep seabed sediments and permafrost areas. Due to their vast reserves, they are regarded as a future energy resource [1,2]. China, the USA, Japan, Canada, and India have conducted hydrate exploration and exploitation in recent decades [3]. However, the hydrate decomposition in the sediments affects mechanical response of hydrate-bearing
sediments [4]. It causes engineering risks, environmental risks, and has climate impacts [5–7]. At the same time, the fluid flow mechanisms and heat transfer characteristics of gas recovery, from the hydrate-bearing sediments (HBS), are complex in deep-water conditions and permafrost areas [8,9].

Nanoscience has been applied in hydrate research, including hydrate-based technologies upstream and downstream, such as core analysis, gas storage, and separation [10,11]. Scanning electron microscopy (SEM) is performed with an electron microscope that produces images of a sample by scanning the surface with a focused beam of electrons. Based on the advanced technology of a low temperature in Cryo-SEM, it can provide hydrate sample surface morphology information and hydrate microscopic properties [10,12,13], such as the sample composition of different particle microstructures and the relationship between the contact surface and pore structure [14–17]. It can also clearly reflect the decomposition process of the hydrate sample surface changes. The surface characteristics, surface microstructure, particle characteristics (size, shape, and surface topography), pore characteristics, and their contact can be obtained from pure hydrate samples, hydrate-bearing sediment samples, and sediment particles. Understanding NGH particle characteristics, pore structure, and other properties will not only help to reveal the physicochemical properties of hydrate and its growth environment but also help to calculate the gas hydrate saturation and correct calculation model of hydrate resources [18]. The effects of particle size and hydrate saturation on hydrate growth habit, pore structures, and the permeability of HBS have been investigated by X-ray experiments and pore-network models [19,20].

The characteristics of GH formation and solid-state deformation have been discussed, and GH has undergone extensive strain hardening and solid-state disproportionation or dissolution under the conditions in its conventional hydrostatic stability field [14]. Some scientists have found the gas hydrate and ice profiles in the Mallik sample to be smooth and dense materials surrounding isolated large pores, while other closely juxtaposed profiles show a highly mesoporous structure. The possible sources of mesoporosity and the sources of mixing ice and gas hydrates have been evaluated, both of which affect the interpretation of the physical property measurements. The image laboratory synthesised ratio of methane hydrate + quartz aggregates by phase proportions is comparable to the sand interval shown by the Mallik material [14,18,21].

Onshore and offshore hydrate trial production tests caused the sand production issues, which restrict future commercial hydrate production [22]. In the sand production experiments of hydrate exploitation, it is mentioned that the decomposition of NGH may have a particular effect on the sediments [23], and it is proposed that NGH particles are equivalent to gravel in the reservoir of high silty-clay, which has the function of retaining mud and sand [22,24–26]. The differences in solid particle transports during hydrate production, conventional oil, and gas production, as well as and drilling, have been analysed [27].

In this paper, we hope to observe the micromorphology of NGH in sediments samples and the microcosmic process of NGH decomposition through Cryo-SEM experiments. Furthermore, the microcosmic sand production characteristics during the hydrate decomposition process in sediments are discussed.

2. Experimental Apparatus and Process

2.1. Experimental Apparatus

To perform the experiments, the type S-4800 Cold Field Scanning Electron Microscope (Cryo-SEM) by Hitachi Co was used. Due to the particularity of the HBS samples, the samples must be kept in low-temperature conditions during sample preparation, delivery, and processing in the chamber (Figure 1). Therefore, the type PP3010T frozen transmission system of Scanning Electron Microscopy (Cryo-SEM Preparation System) by British Quorum Co. (East Sussex, UK) was used for the sample and recycling. The experi-
mental principle is to observe the surface morphology of various solid samples by an electron beam acting on the sample surface. Electron images with a high depth of field morphology contrast or composition contrast of micrometre and nanometre resolution are taken. The main technical indexes of the S-4800 are shown in Table 1.

Table 1. Main technical indexes of the SEM facility.

| Index                        | Key Parameters                                                                 |
|------------------------------|--------------------------------------------------------------------------------|
| Resolution ratio             | 1 nm (15 kV), 2 nm (1 kV)                                                      |
| Acceleration voltage         | 0.5–30 kV                                                                      |
| Amplification factor         | x20–x800000                                                                    |
| 3-axis motor drive sample table: moving range: X:0–50 mm; Y: 50 mm; Z: 1.5–30 mm; T: −5° + 70°; R: 360° |
| Maximum sample size:         | 100 mm                                                                         |

Figure 1. Cryo-SEM and refrigeration transmission system.

2.2. Experimental Material

Six types of mesh were applied to sieve the sand: 100–150 mesh, 150–200 mesh, 200–300 mesh, 300–500 mesh, 500–1000 mesh, and 1000–2000 mesh; the median grain size of the sand were 150 μm, 87 μm, 55 μm, 383 μm, 23 μm, and 13 μm, respectively (Figures 2 and 3). Self-made deionized water and pure methane gas were applied in the experiments. The details of HBS formation are discussed in our previous study [28].
Figure 2. The particle size distribution in different types of mesh.

Figure 3. SEM images of six sizes of grain samples: (A) 150 μm, (B) 87 μm, (C) 55 μm, (D) 38 μm, (E) 24 μm, and (F) 13 μm.

2.3. Experimental Procedure

The Cryo-SEM method is an effective way to observe the microscopic morphology and change characteristics of hydrate sample particles. However, due to the easy decomposition characteristics of the hydrate samples [29], the experimental process was carefully designed as follows:

(1) HBS samples were prepared and obtained by using the quick-opening technology of the liquid nitrogen spray reactors. Then, the samples were stored in liquid nitrogen at
low temperature until tested by Cryo-SEM. (Liquid nitrogen not only prevents the decomposition of NGH, but also isolates the wet air and prevents frosting)

(2) We placed the hydrate bearing-sediment samples, wrapped in tin foil, into the sample table. The above process required pre-cooling and cooling of the operating tools, for which the wrapping materials and the sample table were in a liquid nitrogen environment.

(3) The packed sample table was quickly transported to the pretreatment chamber of the freezing transmission system through the freezing transmission rod. Then, the sample was cut and stripped by a freezing fracture knife in a vacuum condition, revealing a fresh section of the sample. (To avoid the interference of surface condensation, water vapour and frost formation, as well as liquid nitrogen and nitrogen can be extracted under vacuum conditions)

(4) The sample with the fresh section was transferred from the freezing transmission device to the Cryo-SEM sample table. The equipment parameters were observed and controlled. (At this point, it is necessary to avoid the sublimation of the sample in the vacuum test environment and reduce the damage of the electron beam and electrophoresis to the sample surface.)

It is difficult to synthesize methane hydrate in low-permeability sediments, so the multi-shock method was used to synthesize methane hydrate at 275.13 K ~ 278.15 K. After the synthesis, the characteristics of NGH in sediments with different particle sizes were examined, as shown in Table 2.

| Type | Screen (Mesh) | Sediments d50(μm) | Sediment Classification | Initial Pore-fication (%) | Hydrate Saturation (%) |
|------|--------------|------------------|-------------------------|---------------------------|------------------------|
| A    | 100–150      | 150              | fine sand               | 42.11                     | 35.61                  |
| B    | 150–200      | 87               | very fine sand          | 41.91                     | 34.32                  |
| C    | 200–300      | 55               | coarse silt             | 38.13                     | 36.25                  |
| D    | 300–500      | 38               | coarse silt             | 38.66                     | 35.01                  |
| E    | 500–1000     | 24               | medium silt             | 38.04                     | 33.22                  |
| F    | 1000–2000    | 13               | fine silt               | 37.84                     | 32.43                  |

3. Experimental Results and Discussion

3.1. Morphological Characteristics of NGH in Sediments

In Figure 4, the distribution of NGH and ice can be seen in the sediments. The macroscopic occurrence of NGH in the sediments includes lens [30], block, vein, nodular and diffuse.

(1) The microscopic feature of NGH in the sediment highlighted ice crystals and a porous structure after NGH decomposition. The visible pores expanded during methane hydrate decomposition (from Figures 5–9). The distribution of NGH in the sediments included the vein, cluster, block, membrane, and spherical. The cluster is mainly free and partially cemented with sand, while the block was cemented with sand and mostly embedded in the sand.

From the perspective of different grain size distribution, the hydrate occurrence in 100–150 mesh sediments was mainly veined NGH, with obvious fractures between the sediments and veined hydrates in fissures, and the NGH was cemented together with sediments (Figure 5). The hydrates in the 150–200 mesh sediments were mainly clusters and spherical NGH (Figure 6). The hydrates in the 200–300 mesh sediments were mainly massive NGH and NGH blocks embedded in the sediments (Figure 7). There were apparent clusters of hydrates in the 300–500 mesh sediments, as well as hydrate membranes partially wrapped around sediments (Figure 8). In the 500–1000 mesh sediments, most were clusters of NGH and hydrate membranes wrapped around sediments (Figure 9). Therefore, the different particle sizes of sediment have a large influence on the distribution of NGH. The smaller the sediment is, the more contact it has with wrapped and cemented NGH.
It is worth noting that spherical NGH (Figure 4) was found. Some scholars have interpreted this spherical material as marine organisms (there are apparent sharp snout cracks on the spherical material, which are presumed to be feeding locations). In contrast, a similar spherical material has been detected in pure water, but there were no obvious sharp snout cracks [31]. Some scholars suggested this spherical material could be a frozen bubble of dissociated gas in the dissociated water during the sudden temperature drop via liquid nitrogen. However, this spherical material was not seen in the pure sediment system. Therefore, it is speculated to be methane hydrate.

(2) There were many ice morphologies. A distinct hexagonal crystal was visible in this experiment (Figure 4). Since the water, frozen by liquid nitrogen, does not easily form a distinct hexagonal crystal, it is speculated that the hexagonal crystals of about 10 μm were formed in the process of NGH synthesis (Figure 4).

(3) Mud and sand appeared in a variety of morphologies, mainly, as particles.

Figure 4. Methane hydrates and ice in different sediments: (a) spherical and cluster hydrates, (b) porous media during hydrate dissociating, and (c) block hydrates with ice crystals.

Figure 5. SEM observed the evolution of NGH in 100–150 mesh sediments. (a) 0 min, zoom 3.1mm × 1.8k (scale 30 μm), (b) 2 min, zoom 3mm × 2.5k (scale 20 μm), (c) 3 min, zoom 3mm × 2.5k (scale 10 μm) and (d) 5 min, zoom 3m × 11.0k (scale 5 μm).
Figure 6. SEM observed the evolution of NGH in 150–200 mesh sediments. (a) 24 min, zoom 2.9mm × 6.0k (scale 5 μm), (b)27 min, zoom 2.9mm × 18k (scale 3 μm), (c)20 min, zoom 2.9mm × 25.0k (scale 2 μm), (d)23 min, zoom 3m×2.5k (scale 1 μm), (e)0 min, zoom 2.9mm × 25.0k (scale 10 μm) and (f)2 min, zoom 3m×2.5k (scale 5 μm).
Figure 7. SEM observed the evolution process of NGH in 200–300 mesh sediments. (a) 0 min, zoom 4.3mm×4.50k (scale 10 µm), (b) 2 min, zoom 4.3mm×4.50k (scale 10 µm), (c) 4 min, zoom 4.3mm×4.50k (scale 2 µm), (d) 7 min, zoom 4.3mm×10.0k (scale 5 µm), (e) 8 min, zoom 4.3mm×60.0k (scale 0.5 µm), (f) 9 min, zoom 4.3mm×60.0k (scale 0.5 µm), (g) 10 min, zoom 4.3mm×60.0k (scale 0.5 µm), (h) 11 min, zoom 4.3mm×60.0k (scale 0.5 µm) and (i) 12 min, zoom 4.3mm×10.0k (scale 5 µm).

Figure 8. SEM observed the evolution process of NGH in 300–500 mesh sediments. (a) scale 10 µm, (b) scale 5 µm, (c) scale 5 µm, (d) scale 2 µm.
Figure 9. SEM observed the evolution process of methane hydrate in 500–1000 mesh sediments. (a) 0 min, zoom 4.3mm × 15.0k (scale 3 μm), (b) 1 min, zoom 4.3mm × 50.0k (scale 1 μm), (c) 2 min, zoom 4.3mm × 35.0k (scale 3 μm), (d) 3 min, zoom 4.3mm × 15.0k (scale 3 μm), (e) 7 min, zoom 4.3mm × 35.0k (scale 1 μm), (f) 6 min, zoom 4.3mm × 50.0k (scale 1 μm), (g) 11 min, zoom 4.3mm × 15.0k (scale 3 μm), (h) 23 min, zoom 4.3mm × 15.0k (scale 3 μm) and (i) 26 min, zoom 4.3mm × 50.0k (scale 1 μm).

3.2. Characteristics of Microscopic Morphology during the NGH Decomposition Process in Sediments

It can be seen from Figures 5–9, although the NGH was at a low temperature of 77.15 K in liquid nitrogen, it decomposed in a vacuum environment and accelerated its decomposition under the irradiation of electron beams.

It can be seen that an 8 μm base of porous methane hydrate block was embedded into the sediment, and a 5 μm porous methane hydrate was firmly mounted on the top (Figure 7a). Two minutes later, the top methane hydrate block not only shrank due to decomposition but also collapsed due to the decomposition and softening of the connecting base (Figure 7b). After 2 min, the top methane hydrate block fell further and shrank (Figure 7c) until the methane hydrate block fell behind the base after 3 min (Figure 7d). The tip on the base was subtly observed in the next 1 min (Figure 7e–h). The figures show that the tip on the base, over the next 1, 2, and 3 min, gradually disappeared with the NGH decomposition. Finally, the base collapsed (Figure 7i). From Figure 7a,d,i, the continuous evolution...
process of the embedded methane hydrate block and base showed gradual decomposition in just 12 min.

During the decomposition of the NGH (Figure 8a), the distance between clustered methane hydrates gets larger and larger, from 1.25 μm (Figure 8b) to 2.4 μm (Figure 8c). The clustered NGH also became more porous (Figure 8d), and it may leave more space after it is completely decomposed. Thus, this verifies that methane hydrate decomposition increases the pore space of the reservoir. At the same time, the expansion into the space by the decomposition gas or decomposition water would generate a driving force on the surrounding particles.

Methane hydrate, ice, and methane hydrate film-wrapped sediments were obvious (Figure 9a). Zooming in on the sticky area, it can be seen that the sticky area of the NGH was disconnected as it decomposed. Deterioration of the hydrate-ice film interface, which enveloped sediments, was observed (Figure 9b,c). Then, the hydrate-ice film interface degradation of the wrapped sediment gradually expanded (Figure 9d,e), and the degraded surface showed small ice crystal particles (Figure 9f–i). With the further degradation of the membrane interface, the gap between the methane hydrates also changed from the connection: from 1.6 μm to 2.3 μm. It is speculated that the expansion force on the space by the decomposition gas of NGH may provide a driving force for the particles’ migration. It was verified that the pore space between the particles was gradually enlarged during the decomposition of NGH (Figure 9f–i). It is worth noting that, although the NGH film at the interface decomposed and degraded, the ice crystal particles on the muddy surface from the decomposed water gradually increased. If it was a non-low temperature liquid nitrogen environment, the decomposed water would be on the surface of the particles or between the particles. The increase in water content may soften the solid-state stability and cementing strength (Figure 9i).

4. Discussion

Granular material is a discrete system composed of a large number of particles of macroscopic sizes, such as soils and fractured rocks, which is determined by the discrete nature of these media [32–34]. In this research, the hydrate particle size in the actual formation (concentrated at 200 μm) may be larger than the silty-clay (Figure 10). The microscopic features of NGH decomposition in sediments, as well as their effect on sediment migration, are observed and discussed below (Figure 11–13).
Figure 10. Mean crystal size histogram of hydrate in the field (the Black sea, Indian expedition, Gulf of Mexico, and Hydrate Ridge) and the sediments in our tests. (data from [16] and our tests).

Figure 11. The HBS production, in different stages, from laboratory to field. In the laboratory: (a) sediments with gas diffusion, (b) small size of hydrate formation in pore-filling, (c) hydrate formation in grain-cementing, (d) small size of hydrate formation in pore-filling and grain-cementing, (e) large size of hydrate formation in clumping, and (f) hydrate decomposition. In field production: (g) the first stage, (h) the second stage, and (i) the third stage of HBS production.
Figure 12. The mechanism of hydrate formation and decomposition in different grain sizes of sediments: (a) pure sand, (b) hydrate formation in pore-filling, (c) hydrate formation in clumpy, and (d) hydrate decomposition.

Figure 13. The different mechanisms of sand production between (a) a hydrate well and (b) an oil and gas well.
(1) Solid NGH dissipation

In the sediments (Figure 11a), there were three types of HBS: pore-filling (Figure 11b), grain-cementing (Figure 11c), and mixing (Figure 11d). The clumping type (Figure 11e) of HBS was one of the HBS mixing types with NGH growth. NGH decomposition in the sediments led to the elimination of solid NGH and increased the pore space in the reservoir (Figure 11f). Decomposed NGH in the sediments led to the cementing strength decrease in the sediments. Microscopic deformation and the subsidence of the sediments, due to NGH decomposition, are mechanisms for the visible deformation and subsidence of the reservoir under overlying stress (crustal stress).

(2) Decomposed fluids from NGH.

The driving effect of the decomposed NGH on the surrounding particles was observed, which verifies the previous hypothesis about the driving effect of NGH decomposition on the microscopic solid particles (Figures 11g–i and 13a). That is, during the decomposition of NGH:

(2.1) The rapid expansion of the decomposed gas and decomposed water in space and the generation of expansion thrusts may have had a pushing effect on the surrounding solid particles (Figure 13a), while the fluid produced by NGH decomposition contributed to the driving force and displacement of sediment particles.

(2.2) The decomposed water from the NGH may reduce the strength between the particles, soften the reservoir, and reduce the strength of the reservoir.

During the three stages of HBS field production in vertical and horizontal wells [26,35], these properties can lead to sand production, deformation, and subsidence by reservoir particle migration during NGH extraction (Figure 13g–i).

(3) The formation and decomposition of NGH have different effects on sediments with different particle sizes.

Different grain size sediments (Figure 12a) formed different grain size NGH (Figure 12b,c), and the decomposition of NGH with different grain sizes had different effects on sediments with different grain sizes (Figure 12d). With regards to the observed differences in the formation modes of NGH in different sediment sizes in the experiment, some scholars have pointed out that sediment particles would affect the parameters of NGH distribution. The different sizes of sand types A to E can be seen in Figure 12a. It was found that sediment particle size may affect the particle size of hydrate formation, in pore-filling (Figure 12b) and clumping (Figure 12c), during the laboratory synthesis process. The average size of hydrate in the field is about 200 μm (Figure 10), which is much larger than the sand (Figure 12c). Therefore, during the process of hydrate decomposition with different particle sizes, the reservoir sand is affected differently, and the sand production mechanism and sand control mode of the reservoir are affected (Figure 12d).

(3.1) The volume proportion of NGH generated in small grain sediments was relatively large. Small grain sediments have stronger cementing and adhesion to (Figure 12a) NGH.

(3.2) When the hydrate particles were larger than the sediment particles, the hydrate particles acted as “gravel packing” with the effect of sediment blocking, which may affect the sand control design accuracy (Figure 12b,c).

(3.3) Small grain size sediments require a less critical start-up migration force [34], so the force generated by NGH decomposition had a greater impact on small grain size sediments. (Figure 12d).

(4) The unique sand production mechanism of NGH

The sand production mechanism of the conventional petroleum industry (oil well, gas well, and gas well with water) is divided into mechanical failure mechanism (shear destruction, lift damage, and bond damage) and chemical failure mechanism (the contact force between the particles, as well as friction and the bonding force between the particles
and cement) (Figure 13b). However, the decomposition of single NGH particles not only has the above destruction processes but also has sand production through the expansion force caused by the increase in the space volume during the decomposition of NGH (Figure 13a). It is the sand production, caused by the single (partial) hydrate particles that are not seen in the conventional industry, which promotes the migration of solid particles. The sputtering (splashing) effect of NGH on different sizes of types A–F sand is different through the rapid water and gas volume expansion. This provides the driving force for the solid particles’ (hydrate and sand) migration and collision. At the same time, the heat and mass transfer characteristics of HBS are affected by the different sizes of sand and hydrate. With the complex coupling of the thermal-hydrological-mechanical-phase change, the sand production of NGH would differ from the conventional petroleum industry.

5. Conclusions

The low-temperature scanning electron microscope observation of methane hydrate decomposition in sediments not only verified the hypothesis that the decomposition of hydrate particles was different from that of conventional oil and gas engineering but also explained the microscopic characteristics of NGH decomposition in sediments:

1. The solid gas hydrates decomposition increased pore space and decreased reservoir strength. The sand production, deformation, and subsidence of the NGH reservoir can be promoted jointly under the action of water decomposition and gas expansion, as well as the softening of water decomposition and overburden stress (crustal stress).

2. NGH decomposition had a more significate influence on small size sediments, which provides a reference for the selection of hydrate production target areas.

3. The gas hydrate particle size may be larger than the mud sand particle size in the reservoir and act as the “gravel” role, so the gas hydrate particle size cannot be ignored in the sand control design.

4. The complex coupling of the thermal-hydrological-mechanical-phase change in HBS would be affected by the different sizes of sand and hydrate. Rapid volume expansion, by decomposed gas and water from NGH, caused the solid particle sputtering (splashing). This is a unique process of sand production in gas hydrate wells, which is different from that in gas and oil wells.

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