# Current status and progress of research on hydrate-bearing sediments

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#### Abstract

The exploitation of hydrate-bearing sediments faces many challenges, such as geotechnical problems, difficulties in reservoir physical property analysis, and phase equilibrium issues. In terms of geotechnical aspects, the artificial preparation of largescale hydrate-bearing sediment samples is extremely difficult, and the fine detection and quantitative characterization of micro-meso fabric are also full of challenges. In reservoir physical property analysis, although low-field nuclear magnetic resonance technology has advantages, it has low quantitative accuracy and high implementation cost. The phase equilibrium relationship is significantly affected by sediment physical properties, pore water salinity, and gas components. Moreover, there are difficulties in sample preparation methods and theoretical models. Existing exploitation technologies include depressurization mining and solid fluidization mining. The former is prone to cause problems and affect efficiency, while the latter has made breakthrough progress. The research progress of exploitation technologies shows the characteristics of multitechnology integration and the progress of high-resolution imaging technology. Innovative directions include the application of low-field nuclear magnetic resonance technology and multi-field coupling numerical simulation. Despite challenges, through technological innovation and multidisciplinary cooperation, safe and efficient exploitation is expected to be achieved, providing a new potential way for global energy demand.

#### **Keywords**

Hydrate-bearing sediments; exploitation challenges; low-field nuclear magnetic resonance technology; multi-field coupling numerical simulation.

# 1. Introduction

Natural gas hydrate as a popular emerging source in this century is very rich in reserves. According to statistics, globally detectable reserves of natural gas hydrate compounds are about  $2 \times 1014$ m3, containing a total of  $1 \times 1011$ t of organic carbon content, which is about twice the carbon content of fossil energy sources (coal, oil, and natural gas) that have been discovered so far <sup>[1,2]</sup>. The environmental conditions of low temperature and high pressure are critical for the generation of gas hydrates, so the general distribution of gas hydrates in nature occurs in permafrost zones and deep-sea bottoms. As shown in Figure 1, less than 3% of terrestrial gas hydrates and more than 97% of gas hydrates belong to hydrate-bearing sediments <sup>[3]</sup>.

Since the 1990s, after a long time of exploration and scientific research, people have found a large amount of natural gas hydrate mineral reserves in the Qilianshan tundra in the Shenhu Sea area of the South China Sea, and the reserves are so huge that they can be widely used in the future <sup>[5,6]</sup>. After continuous theoretical research and field surveys, China achieved the first successful test mining of natural gas hydrate in the South China Sea's Shenhu Sea in 2017, and the proven reserves of natural gas hydrate deposits in the South China Sea waters total eighty billion tons of oil equivalent, which is one-half of the total proven oil and natural gas deposits.

The development and utilization of natural gas hydrate resources in the South China Sea is an indispensable part of realizing China's strategy of ocean power and ensuring energy security. China has conducted two consecutive natural gas hydrate test mining in the Kobe Sea area of the South China Sea, and has maintained a number of world records <sup>[7]</sup>.

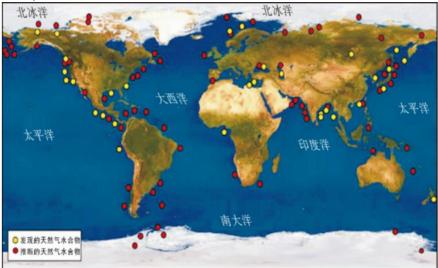


Fig.1.1. Map of proven gas hydrate distribution [4].

However, in-situ extraction of natural gas hydrate is a complex multi-field coupling process affected by multiple factors, accompanied by temperature, hydraulic pressure, mechanical and chemical coupling of material phase change, seepage and skeleton deformation, etc. This process will lead to the deterioration of hydrate-containing sediments, triggering a series of oceanic geotechnical hazards, such as continental shelf landslides, seabed collapse, etc. <sup>[8]</sup>. In addition, failure to control the dissolution rate of natural gas hydrates will lead to the escape of large amounts of methane gas into the atmosphere, thus exacerbating the greenhouse effect <sup>[9]</sup>.Despite the many challenges, the exploitation of hydrate-bearing sediments is of great significance. It offers new potential ways to address global energy needs and is expected to occupy an important place in the future energy mix. With the continuous advancement of technology, it is expected that the difficulties in the mining process can be gradually overcome and safe and efficient mining can be realized through the combined use of traditional techniques and modern analytical methods, such as low-field nuclear magnetic resonance, X-ray diffraction and computed tomography.

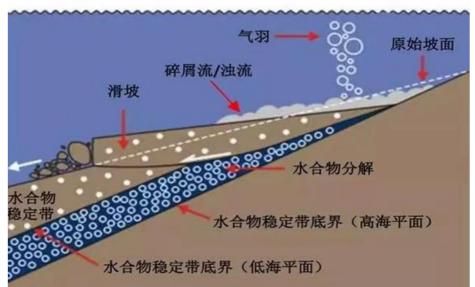


Fig.1.2. Hydrate decomposition-induced landslides on slopes[10].

# 2. Challenges to the mining of hydrate-bearing sediments

#### 2.1. Soil mechanics issues

The manual preparation of large-scale hydrate-bearing sediment samples faces a number of difficulties. On the one hand, natural gas hydrates are usually found in complex environments such as seafloor sediments and permafrost zones, and obtaining large-scale raw sediment samples is a great challenge in itself. For example, when sampling on the seafloor, one needs to overcome the huge water pressure, complex marine environment and high sampling cost. On the other hand, it is not easy to simulate the preparation of large-scale hydrate-bearing sediment samples in the laboratory. It is often difficult to accurately restore the physical and mechanical properties of sediments and the distribution of hydrates in the natural environment with the current preparation methods. According to related studies, the saturation of hydrates in in situ soil is high after a long period of geological action, but the solubility of methane gas is too low, and the limitation of the water-gas interface makes hydrate synthesis extremely slow. When preparing hydrate-containing soils indoors using the saturation method, it is difficult to synthesize specimens with the same high degree of saturation as in situ. For example, Wei Changfu's research team at the Wuhan Institute of Geotechnics, Chinese Academy of Sciences, developed a gas hydrate spray synthesis device, which facilitates the preparation of pure hydrates by spraying a water mist in a high-pressure, low-temperature environment so that the gas can rapidly synthesize a hydrate powder upon contact with the water mist, but there is still a long way to go in terms of large-scale specimen preparation.

#### 2.2. Phase equilibrium problem

Sediment physical properties, pore water salinity and gas components all have significant effects on hydrate phase equilibrium relationships. Firstly, the physical properties of sediments can change the conditions of hydrate generation and decomposition. Different sediment particle sizes, porosities and other physical characteristics affect the stability of hydrates in sediments. For example, finer sediment particles may provide more surface area, which is favorable for hydrate formation, but may also lead to pore blockage, affecting gas diffusion and hydrate decomposition. Second, pore water salinity also has an important effect on hydrate phase equilibrium. Higher salinity decreases the stability of hydrates and shifts the phase equilibrium conditions toward higher pressure and lower temperature. It has been shown that the decomposition temperature of natural gas hydrates increases significantly with increasing pore water salinity in the laboratory. In addition, gas components also affect the phase equilibrium conditions. For example, the phase equilibrium curves of methane hydrate and propane hydrate are very different.

In the study of phase equilibrium relationships, there are many difficulties in both sampling methods and theoretical modeling. In terms of sampling methods, the generation and decomposition of natural gas hydrates in sediments in nature are very different from those in pure water systems, and it is very difficult to accurately simulate the phase equilibrium conditions of hydrate sediments in natural environments in the laboratory. Current sampling methods are often difficult to simultaneously consider the effects of various factors such as sediment physical properties, pore water salinity and gas components, resulting in low quantitative accuracy of sampling. For example, in some experimental studies, although many advanced methods and technologies have been applied, there is still the problem of not being able to accurately control the physical properties of sediments and the salinity of pore water. In terms of theoretical models, most of the current hydrate sediment phase equilibrium theoretical models are improved from the van der Waals-Platteeuw (vdWP) statistical thermodynamic model, which generally requires the provision of microscopic pore size

distribution parameters as input conditions, and the calculation process is extremely complex. The complexity of the expressions, the large number of parameters, and the difficulty in obtaining them limit their application in natural gas hydrate extraction. For example, the temperature-pressure equilibrium conditions calculated by the vdWP model differ greatly from the macroscopic test results. In addition, in real hydrate sediments, even if the gas is sufficiently abundant and the temperature-pressure of the sediments meets the hydrate generation conditions, not all the water in the sediments can be converted into hydrates, and some liquid water still coexists stably with the hydrates. Previously, no model has considered the dependence of the liquid water content in sediments on temperature, pressure, and concentration, which has brought great challenges to the study of phase equilibrium relationships.

# 3. Innovative directions for mining hydrate-bearing sediments

# 3.1. Application of low-field nuclear magnetic resonance technology

Low-field nuclear magnetic resonance technology plays a crucial role in analyzing reservoir pore structure and fluid distribution during hydrate mining. By detecting the magnetic resonance signal of hydrogen nuclei in the sample and analyzing its transverse relaxation time (T2) distribution, pore sizes and fluid types in the reservoir can be identified in detail. For example, in argillaceous silt-type natural gas hydrate reservoirs, this technology can quantify the changes in pore structure under the action of clay minerals and provide data support for formulating mining strategies. During the formation process of hydrates, low-field nuclear magnetic resonance technology can monitor changes in sediment pore size and distribution. For example, as hydrates are generated, the maximum pore radius and average pore radius of the sediment aqueous phase pore space gradually decrease, and the fractal coefficient of the pore space gradually increases. This helps to deeply understand the occurrence state of hydrates in sediments and provides a scientific basis for the location and mining of hydrates.

Low-field nuclear magnetic resonance technology is of great significance in real-time monitoring of hydrate formation and decomposition. In the laboratory synthesis and mining simulation process of hydrates, this technology can monitor the formation and decomposition process of hydrates in real time. This function is crucial for studying the kinetic characteristics of hydrates and optimizing mining conditions. Through real-time monitoring, we can understand the changes of hydrates under different temperature and pressure conditions, adjust mining parameters in time, and improve mining efficiency and safety. For example, in the stepwise depressurization mining experiment of natural gas hydrates, low-field nuclear magnetic resonance technology can monitor the decomposition of hydrates in real time and help researchers control the depressurization rate, effectively reduce water production and improve gas recovery rate. In addition, combined with triaxial experiments, the mechanical properties and internal damage evolution characteristics of hydrate-bearing sediments under different loading conditions can also be analyzed to further evaluate the stability and safety of the reservoir during the mining process.

# 3.2. Multi-field coupling numerical simulation.

When constructing a macroscopic multi-field coupling model, multiple factors need to be considered. First of all, it is necessary to accurately describe the thermophysical properties of hydrates, including parameters such as thermal conductivity and specific heat capacity. These parameters will affect the temperature change of the reservoir. Secondly, the simulation of fluid flow is also crucial. The flow characteristics of water, natural gas, and gas generated by hydrate decomposition in the reservoir need to be considered. In addition, in terms of solid mechanics, the deformation and stress distribution of the reservoir should be considered. Because the

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decomposition of hydrates will lead to changes in the effective stress of the reservoir, which may cause problems such as formation deformation and sand production.

The mesoscopic sand production model plays an important role in studying the sand production law. By simulating the depressurization mining process through a macroscopic multi-field coupling model, data such as effective stress, hydrate saturation, and water-gas flow velocity can be obtained. These data are transferred as boundary and initial conditions to the mesoscopic sand production model for dynamic sand production simulation. The mesoscopic sand production model can study the occurrence and development process of sand production from a microscopic perspective and clarify the sand production mechanism and law. For example, a hydrate sand production model constructed based on the discrete element method can consider core elements such as hydrate occurrence mode, hydrate decomposition effect, and fluid-solid coupling. In terms of hydrate occurrence mode, different occurrence modes will affect the decomposition process and sand production behavior of hydrates. The hydrate decomposition effect will change the physical properties of the reservoir and increase the possibility of sand production. The fluid-solid coupling effect considers the interaction between fluid flow and solid deformation, which is crucial for accurately predicting the sand production law. By combining the macroscopic multi-field coupling of heat-fluid-solid and the mesoscopic sand production model, the sand production problem in the mining process of hydrate-bearing sediments can be studied more comprehensively, providing important theoretical support for scientifically designing sand control and mud control completion methods and promoting the industrialization process of hydrates.

# 4. Conclusion

This paper provides a comprehensive review of mining hydrate-bearing sediments. It is shown that the mining of hydrate-bearing sediments faces many challenges, including geomechanical problems, difficulties in analyzing reservoir properties, and phase equilibrium problems. In terms of geotechnical problems, the manual preparation of large-scale hydrate-bearing sediment specimens is difficult, while the fine detection and quantitative characterization of micro- and microstructures are also challenging. In the analysis of reservoir physical properties, low-field nuclear magnetic resonance (NMR) technology has the advantages of low equipment cost, rapid analysis and testing, high accuracy, no loss of samples, and simple sample preparation, but it also has the problems of low quantitative accuracy and high implementation cost. In the study of phase equilibrium relationship, sediment physical properties, pore water salinity and gas components have significant influence on hydrate phase equilibrium relationship, and there are difficulties in sample preparation methods and theoretical models. In the direction of innovation, low-field nuclear magnetic resonance technology provides key support for pore structure analysis and real-time monitoring for hydrate mining; multi-field coupled numerical simulation provides important theoretical support for the study of sand release mechanism by combining the heat-fluid-solid macroscopic multi-field coupling and fine-scale sand release model.In conclusion, although the exploitation of hydrate-containing sediments faces many challenges, it is expected to gradually overcome them through continuous technological innovation and multidisciplinary cross-cooperation to realize safe and efficient exploitation and provide a new potential way for global energy demand. In the future, we should continue to increase our research investment in hydrate-containing sediment mining technology, explore more advanced and efficient mining methods, and contribute to the sustainable development of the energy sector.

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