Accumulation model of gas hydrate in the Chaoshan depression of South China Sea, China

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Abstract. The northern waters of the South China Sea are rich in natural gas hydrate resources. The source of gas is a crucial factor controlling the formation of natural gas hydrates. Gas sources that form natural gas hydrates can be related to three categories: pyrolysis genesis, biological genesis, and inorganic genesis. A series of large- and medium-sized Mesozoic and Cenozoic sedimentary basins have developed in the northern South China Sea. These basins are rich in organic matter and there are multiple sets of source rocks buried deeply, which are the main gas sources for hydrate accumulation. The Chaoshan Depression is the largest Mesozoic residual depression in the northern South China Sea. Analysis of geochemical and geological samples show that there is a significant oil and gas leakage activity on the seabed. Seismic data and drilling confirmed the existence of natural gas hydrates in Chaoshan Depression. The natural gas hydrate accumulation pattern of Chaoshan Depression is a leaky accumulation, which has the characteristics of unique structure, abundant Mesozoic gas source, and development of faulting system.

1. Introduction
Natural gas hydrate, also known as combustible ice, is a solid substance formed by water and natural gas (mainly methane) under low temperature and high pressure conditions. The primary component of gas hydrate is methane and water \cite{1-2}. Gas hydrate show various occurrences, such as massive, vein, nodule and dispersed \cite{3-6}. There are three genetic models of gas hydrate reservoirs, i.e., diffusive model, seeping model and complex model in South China Sea \cite{7-10}. Gas hydrate is an unconventional energy source with great potential on the earth \cite{11-12}.

The gas source is an important control factor for the formation of natural gas hydrates. According to the different generation mechanisms, the gas sources that form natural gas hydrates can be subdivided into three categories: pyrolysis genesis, biological genesis and inorganic genesis. Pyrolysis genesis is the most important gas sources. In the northern part of the South China Sea, large and medium-sized Mesozoic and Cenozoic sedimentary basins such as the Pearl River Mouth, Qiongdongnan, Xisha Trough, Shuangfeng, have developed. The deposition rate of those basins is large, and the maximum deposition thickness can reach 11 km. These basins are rich in organic matter, and there are multiple sets of source rocks buried deeply. Many large and medium-sized oil and gas
fields have been discovered, showing a good source of pyrogenic gas. The stable area of natural gas hydrate in Shenhu sea area is mainly located in Baiyun Sag, with a water depth of 200-2000 m. In the deeper part of Baiyun Sag, there are huge thick Paleogene source rocks, forming a large number of oil and gas fields such as LH19-3-1, LH19-1, PY30-1, PY34-1, PY29-1 and PY35-1 [13]. Leakage is an important source of natural gas hydrate in the shallow area of Shenhu [2].

The accumulation characteristics and genetic models of gas hydrate reveal three gas hydrate reservoirs are developed in the NE slope of the South China Sea, and their accumulation systems are independent, but genetically related [14]. Diffusive model presents lamellar distribution at the hydrate stability zone, and its bottom boundary is coincident with the distribution of bottom simulating reflector (BSR). Seeping model presents sediment cracks or fractures in the form of block and constitutes multiple mineral ores in various parts of the stability zone. The complex model has the accumulation characteristics of both diffusive and seeping model [10].

The research on coexistence between gas hydrates and deep-water petroleum systems has great significance to exploration and exploitation of both deep-water hydrocarbon reservoirs and shallow gas hydrates[15]. The contributions of deep-seated oil and gas to gas hydrate field have been studied [16]. At present, there are more studies on understanding the gas sources for oil and gas reservoirs in Cenozoic sedimentary basins, but limited research on the fluid sources for oil and gas reservoirs in Mesozoic sedimentary basins. The Mesozoic is an important field of oil and gas exploration and development across the world. The petroleum resources can account for 80.4% of the global total resources, of which marine oil and gas resources of the Cretaceous and Jurassic can account for 54% of the global total resources [17]. Therefore, the study of Mesozoic gas source is especially important. In this paper, the controlling effects of Mesozoic oil and gas reservoirs in the northern South China Sea (Chaoshan Depression) on hydrate accumulation was investigated using findings from studies on the Chaoshan Depression in recent years[18-22]. Further, accumulation model of gas hydrate was established to guide the exploration and development of gas hydrate in China.

2. Geological settings

2.1. Geologic features of Chaoshan depression

The Chaoshan depression is located in the southeast part of the Pearl River Mouth Basin, northern South China Sea (Figure 1), with an area of more than $3.7 \times 10^4$ km$^2$ and a water depth ranging from 300m to 2000m. The Chaoshan depression is a residual Mesozoic depression, which mainly experienced four stages: (a) the Middle-Late Jurassic subsidence, (b) the Late Jurassic to the Early Cretaceous uplift, (c) superimposed burial of the Early to the Late Cretaceous, (d) and then the Late Cretaceous uplift and erosion (Figure 2).

In the Early Jurassic, the Chaoshan depression was mainly filled with shallow-bathyal sediments, and bathyal sediments developed in the center of the depression. The depression undergone a seawater invasion from the southeast direction, and had a main NW and NE source direction advancing toward the center of the depression (Figure 2a).

In the Middle Jurassic, the water depth was the shallowest due to a regression, and the Chaoshan depression was mainly filled with the littoral-neritic facies sediments. Affected by the coastal Pacific domain, the seawater entered from the southeast direction, and the shallow sea sediments developed in the center of the depression; the source direction was mainly the NW and NE directions, it pushed forward to the center of the depression and the source of material was relative sufficient (Figure 2b). The Late Jurassic was also influenced by the coastal Pacific domain, with the increasing water depth and the crustal subsidence, which caused the filling of neritic-bathyal sediments in the Chaoshan depression. The seawater entered from the southeast direction and developed bathyal-pelagic sediments at the center of the depression; the source direction was Mainly the NW and NE directions, and the source supplies were the most abundant (Figure 2c).
At the beginning of the Early Cretaceous, the sedimentary environment on the north area of the Chaoshan depression suddenly changed into the paralic environment due to tectonic uplifting, but the center of the Chaoshan depression and the south area of the depression were still in the bathyal-pelagic
environment. At the late stage of the early Cretaceous, on the north of the Chaoshan depression, a series of littoral-neritic facies sand-mud stones sandwiched limestone and volcaniclastic rock are developed. The seawater withdrew from the northern area of the Chaoshan depression and formed a forced regression deposit towards the ocean basin on the northern slope of the Chaoshan depression. In the late Cretaceous, sedimentary environment transitioned from land to coastal, shallow sea, the north area of the depression center was in the terrestrial facies environment, along with the development of rivers and alluvial plains, lacustrine facies and shoreline delta facies, the central and southern of the Chaoshan depression were in the transitional facies environment (Figure 2d).

Drilling data confirmed the development of marine Mesozoic gas hydrate in the northeastern part of South China Sea. For example, the CFC-1 well drilled the early Cretaceous marine strata in the middle uplift belt of the Taixinan Basin [23]; the LF35-1-1 well drilled in the Middle-Late Jurassic marine sediments and early Cretaceous paralic-facies sediments on the northern slope of the Chaoshan depression [18-22,24-33].

2.2. Source rock condition in Chaoshan depression

In the northern slope belt of the Chaoshan depression, there is only one well drilled by China National Offshore Oil Corporation (CNOOC), named LF35-1-1. The well reveals that the top part of the Middle-Upper Jurassic is gray-black lamellar mudstone and argillaceous siltstone sandwiched siliceous rock, containing a small amount of muddy limestone; while the lower part is gray-black lamellar mudstone and muddy siltstone sandwiched by sandstone and limestone. Mudstones are rich in organic debris. The Cretaceous is generally a set of river-lake sediments. Its upper part is a combination of purple mudstone, siltstone and sandstone with a small amount of muddy limestone, and the lower part is a combination of gray-grained bedded mudstone, siltstone and sandstone with some organic debris.

The geochemical analysis results show that there were two sets of main source rocks (Figure 3), with all located in the Middle-Upper Jurassic, mainly distributed in the intervals of 1940-2022 m and 2100-2400 m. The cumulative thickness of source rocks in the 1940-2022 m interval is 82.28 m, the total organic carbon (TOC) content is between 0.18% and 1.15%, with an average of 0.67%. The cumulative thickness of the source rocks in the wells from 2100 m to 2400 m is 46.16 m. The TOC is between 1.00% and 1.48% with an average of 1.32% [34]. The kerogen types of these two source rocks are mainly type III, and a few of which are type II2. The monostromatic thickness of source rocks is at least 6 m above, and the maximum is more than 40 m, with an average of about 20 m. The average monolayer thickness of the lower source rocks is greater than the upper one. The results of the study indicate that the amount of mudstone in the untouched formation below the well increases significantly. Therefore, the lower shallow marine deposits may have better hydrocarbon generation capability.

The cross-section shows that the Mesozoic in the Chaoshan depression can be roughly divided into the Cretaceous and Jurassic (Figure 4). The source rocks drilled by LF35-1-1 well also penetrated a number of mudstone intervals to the southeast of the depression, and the transgression came from the southeastern part of the structure. The southeast part of the section has more space to accommodate the hydrocarbon sources. The source rocks of the LF35-1-1 well drilled are located in the high system tract (HST), which are some granodiorites. However, this does not mean that the sequence has no hydrocarbon generation potential. Analysis of sequence stratigraphy indicates that there is a sensible phenomenon of onlap in the stratigraphic sequence. Obviously, the accommodation space in the southeast of the basins shows a rapid expansion, which is conducive to the development of source rocks. At the same time, because the LF35-1-1 well is located on the high part of the Chaoshan depression edge. Towards the center of the depression, it is estimated that the number of effective source rock will increase and most of the strata are developed with good source rocks.
Figure 3. Two sets of source rocks in the study area.

The thickness distribution of mudstone in the Chaoshan depression was calculated based on the sand-mud ratio map and the strata thickness map determined by the seismic velocity. The results show that the area with a thickness of > 1600m is mainly distributed in the center part of the Chaoshan depression, showing the NE-SW trend. The thickness of the Jurassic mudstone in the western sag is relatively large, and its value can reach more than 2400 m (Figure 5). Due to proximity the provenance, the source rock of this type has a relatively high organic matter abundance, and the quality of the source rock is also relatively good.

3. Controlling factors for gas hydrate accumulation

The main control factors of hydrate enrichment include tectonic environment, gas source, gas migration control factors, and temperature and pressure conditions. When the temperature is lower than 10°C and the pressure is higher than 10 MPa, the gas hydrate stability zone can be formed.

3.1. Structural framework

The accumulation of gas hydrate are closely related to mass transport deposits (MTDs) and mud diapir structure, which provide a large-scale structural space for hydrate occurrence. MTDs have chaotic facies on the seismic profile and develop during episode of slope instabilities. It involves the remobilization of sediments above the basal shear surface under the influence of gravity. MTDs are often developed on the slope break or steep slope. On the one hand, MTDs structure is favorable for hydrate formation, it is caused by events such as earthquakes, volcanic eruptions, storm waves and rapid accumulation of sediments, or by excessive inclinations of slopes. Loose and organic rich MTDs cause a large number of fluid emissions due to lateral compaction, and gas diffuses and percolates to the shallow part to form hydrate; on the other hand, it is also beneficial for the formation of hydrate, the MTDs may also be the product of hydrate decomposition.

Diapir structure is formed by the material in the deep part of the earth arching or penetrating to the shallow part. Due to the abnormal high temperature and pressure of the formation of diapir, diaper structure constitutes a special type of vertical episodic hydrocarbon expulsion and hydrate accumulation of source rock. When a large amount of fluid moves to the stable temperature and pressure domain required for hydrate formation, gas hydrate can be formed. In the northern slope area of the South China Sea, structural geological bodies such as MTDs and mud diapirs are widely developed, which have a large scale and provide a considerable structural space for hydrates. The extensional faults in the slope area provide a good channel for the vertical migration of deep fracture gas [35].
3.2. Gas source
There are three types of gas sources of gas hydrate: pyrolysis gas, biogas and mixture gas. Pyrolysis gas comes from the pyrolysis of deep source rock, biogas comes from the biogenic gas produced by the degradation of organic matter in the sediment. The mixture gas contains both pyrolysis gas and biogas. The Cenozoic sedimentary basins, such as Qiongdongnan Basin, Pearl River Mouth Basin, Xisha Trough basin and Taixinan Basin, and the Mesozoic residual depression, such as Chaoshan depression have good hydrocarbon generating ability. The huge source rocks are the main substance source [2]. The deep fracture gas is an important source of gas hydrate and shallow gas reservoir. The natural gas leakage from the seafloor is a wide spread natural phenomenon in the world. The gas leakage system in the deep water area is a suitable place for hydrate formation. In the appropriate sea floor environment (temperature and pressure), part of the leaked natural gas will precipitate into hydrate [7], which shows that the hydrate area is accompanied by obvious natural gas leakage. Drilling results show that deep thermal gas is an important gas source of hydrate.

3.3. Controlling factors of gas migration
The controlling factors of gas migration include fracture channeling, fracture leakage and diapirism of mud volcano. The gas source channel of the continental slope in the northern South China Sea is mainly fault channeling. Influenced by the expansion of the central basin in the South China Sea, the continental slope area is in the state of tensile stress, forming a large number of deep and large normal faults, which are connected with the deep hydrocarbon source rock and extend to the bottom of the sea, with good transport capacity, Natural gas from basin pyrolysis can be transported vertically and form hydrate [7] [10] [36-37].

Gas venting is a widely distributed natural phenomenon in the world. In the appropriate submarine environment (temperature and pressure), part of the leaked natural gas will be precipitated into hydrate. Cold spring carbonate rock, chemical auto trophic organism, natural gas hydrate, undersea natural gas overflow, etc. mark the development of undersea natural gas leakage system [38-40].

4. Results

4.1. Evidence of hydrocarbon leakage in the Chaoshan Depression
There are two forms of seepage in subsurface oil and gas reservoirs: macro seepage and micro seepage [41]. Macro-leakage is mainly the migration of hydrocarbons from the reservoir through the overburden and vertical migration to the seabed. Hydrocarbons in subsurface oil and gas reservoirs migrate vertically from the reservoir through the fault systems in the overburden to the seabed sediments, seawater, and sea surface atmosphere, commonly known as micro-leakage of hydrocarbon. This micro-leakage will produce a series of physical and geochemical effects on the seabed sediments. Geochemical exploration of headspace light hydrocarbons, acid-hydrolyzed hydrocarbons, pyrocarbons, altered carbonates, condensed-ring aromatic hydrocarbons, three-dimensional fluorescence, acid-hydrocarbon methane stable carbon isotopes, microbiological testing and analysis etc. were performed using 698 submarine surface sediment samples from the Dongsha Sea. After normal standardization, an abnormal plane distribution map is formed according to the characteristics of abnormal probability distribution (value >1, 12% of the samples). The results show that there are 3 obvious geochemical anomalies in this area (Figure 6), overlapping petroleum reservoir and distribution of BSR. This indicates that there is obvious micro-leakage of hydrocarbons in this area.
The high-resolution three-dimensional bathymetric map of the seafloor shows that the submarine chimney in the Dongsha sea area is very well developed (Figure 7). The results of subsea geological sampling at the S202 station in the Dongsha sea area showed that there were abundant biological shells in the samples (Figure 8), which was the result of bioaccumulation caused by organic gas leakage from the seabed. It can be speculated that there is macro-leakage of oil and gas in the sea area of Chaoshan Depression.
4.2. Geophysical evidence of gas hydrate
The seismic wave velocity of the natural gas hydrate-bearing formation will increase significantly, while the underlying hydrate sedimentary layer will have a significant decrease due to free gas or water content [42-43]. Based on the difference in impedance between the hydrate layer and the underlying stratum, a strong amplitude difference is formed, resulting in a Bottom Simulating Reflector (BSR) appearing on the seismic profile (Figure 9). BSR is a commonly used and most intuitive means of identifying submarine natural gas hydrates, and the distribution of BSR is the main target of exploration and the main basis for resource evaluation [44]. When there is abundant free gas under the hydrate ore belt, an obvious physical interface is formed between the hydrate ore belt and the free gas. The interface can generate strong reflections, showing characteristics such as near parallel to the sea floor, abnormal velocity, reversed polarity, high amplitude, and oblique crossing with the stratum. Obvious BSR can be seen on the seismic profile of Chaoshan Depression (Figure 9).

4.3. Drilling of gas hydrate in Chaoshan depression
In 2019, Guangzhou Ocean Geological Survey Bureau Marine Geology No. 10 drilled hydrates below 60m in the northern seafloor of the Chaoshan Depression (Figure 10). Since the Cenozoic does not yet have the conditions for hydrocarbon formation and the biogenic foundation is poor, the hydrate gas source should come from the lower Mesozoic, and the fracture is an important channel.

5. Discussion
5.1. Distribution and tectonic environment of gas hydrate in Chaoshan Depression
According to the BSR tracking interpretation of seismic data, hydrates in the Chaoshan Depression are mainly distributed in the southwest and middle of the depression (Figure 6). There is a good relationship with the petroleum-bearing structures in Chaoshan depression. This shows that hydrates are affected by oil and gas reservoirs.
Tectonic geological bodies such as slumps and mud diapirs developed in the northern slope area of the South China Sea are widely distributed on a large scale, providing a large-scale structural space for hydrate accumulation (Figure 11). The extensional faults in the continental slope area provide a good drainage system for the vertical migration of deep cracked gas and the lateral migration of shallow biogas.

5.2. Gas source for gas hydrate formation

There are three main types of gas hydrate gas sources: pyrolysis gas, biogas and mixed gas. The gas source of hydrate in Chaoshan depression is mainly pyrolysis gas. Pyrolysis gas originates from the pyrolysis of deep source rocks. As mentioned above, the depression has good hydrocarbon generation capacity, and the thick Jurassic source rock is the main material basis for gas hydrate formation.

The gas source channels of the shallow natural gas reservoirs on the northern slope of the South China Sea are mainly fault channelization. Affected by the expansion of the central basin of the South China Sea, the continental slope area is in a state of tensile stress, forming a large number of deep and large normal faults. These extensional faults communicate with deep source rocks and extend all the way to the bottom of the sea. They have good transport capabilities and can vertically transport the pyrolysis natural gas of the basin below the hydrate reservoirs (Figure 12).
5.3. Model of gas hydrate accumulation

The formation of hydrate reservoirs in the Chaoshan Depression is characterized by the leakage type. The prominent feature is the obvious gas leakage channel in the seismic profile (Figure 11). The combination of the strong reflection and the same-phase axis on the top of the gas chimney and the flanks indicates the hydrate layer. The faults connecting the source rocks and the shallow sedimentary system in the continental slope in the depression are developed well. The gas moves along the fault and unconformity surface from the lower gas source high-pressure region to the upper low-pressure region laterally, or form an ascending flow vertically (Figure 12). When entering the shallow sedimentation, although the pressure and temperature of this upwelling drop, a large number of north-east small and wide faults distributed in the shallow part become the main channel for the fluid to continue to move upward. When the ascending flow of hydrocarbon-rich gas enters the hydrate stability zone, natural gas hydrate can be formed.

The gas source of hydrate comes from deep. The reservoir is muddy sand or fine sand and silt. The gas migrates through the pores, micro-cracks and interlayer faults of the sediment, and is driven by diffusion under the drive of concentration difference, pressure, and capillary force [45] [10] [3]. When the concentration of dissolved methane in the pore water exceeds the thermodynamic equilibrium saturation solubility of the water-hydrate two-phase system, the dissolved methane precipitates to form a hydrate [34], which accumulates as a hydrate at the bottom of the stable domain (Figure 13).

6. Conclusion

1. The Chaoshan Depression is the largest Mesozoic residual depression in the northern South China Sea. Oil and gas leakage from deep reservoirs can form natural gas hydrates.
2. The main controlling factors of hydrate enrichment include tectonic environment, gas source, factors controlling gas migration, temperature and pressure conditions.
3. Geochemical and geological sampling results show significant oil and gas leakage activity on the seabed. Seismic data and drilling confirmed the existence of natural gas hydrates in Chaoshan Depression.
4. The natural gas hydrate accumulation pattern of Chaoshan Depression is a leak-type accumulation, which has the characteristics of unique structural background, abundant Mesozoic gas source, and development of faulting system.

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