

Review Article

Gas Hydrate Accumulation and Occurrence Associated with Cold Seep Systems in the Northern South China Sea: An Overview

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Received 11 March 2021; Revised 29 June 2021; Accepted 30 August 2021; Published 5 October 2021

Academic Editor: Jinze Xu

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Studying deep-water cold seep systems is of great significance to gas hydrate exploration due to their close relationship. Various cold seep systems and related gas hydrate accumulations have been discovered in the northern South China Sea in the past three decades. Based on high-resolution seismic data, subbottom profiles, in situ submergence observations, deep drilling and coring, and hydrate gas geochemical analyses, the geological and geophysical characteristics of these cold seep systems and their associated gas hydrate accumulations in the Qiongdongnan Basin, the Shenhu area, the Dongsha area, and the Taixinan Basin have been investigated. Cold seep systems are present in diverse stages of evolution and exhibit various seabed microgeomorphic, geological, and geochemical features. Active cold seep systems with a large amount of gas leakage, gas plumes, and microbial communities and inactive cold seep systems with authigenic carbonate pavements are related to the variable intensity of the gas-bearing fluid, which is usually derived from the deep strata through mud diapirs, mud volcanoes, gas chimneys, and faults. Gas hydrates are usually precipitated in cold seep vents and deeper vertical fluid migration pathways, indicating that deep gas-bearing fluid activities control the formation and accumulation of gas hydrates. The hydrocarbons collected from cold seep systems and their associated gas hydrate reservoirs are generally mixtures of biogenic gas and thermogenic gas, the origin of which is generally consistent with that of deep conventional gas. We also discuss the paragenetic relationship between the gas-bearing fluid and the seafloor morphology of cold seeps and the deep-shallow coupling of gas hydrates, cold seeps, and deep petroleum reservoirs. It is reasonable to conclude that the deep petroleum systems and gas-bearing fluid activity jointly control the development of cold seep systems and the accumulation of gas hydrates in the northern South China Sea. Therefore, the favorable areas for conventional oil and gas enrichment are also prospective areas for exploring active cold seeps and gas hydrates.

1. Introduction

Cold seeps are seafloor manifestations of a gush or seepage of gas-bearing fluid migrating from beneath the seafloor into the seabed and finally into the water column. They are accompanied by a type of local abnormal sea water with various shapes, such as plumes, columns, and whips, which differ from the physical properties of the surrounding sea water [1–7]. A series of geological, geophysical, geochemical, and

biological processes occur within a cold seep and the surrounding seawater, which are known as a cold seep system or a submarine methane seep system [8–12]. Generally, the activity of a cold seep is episodic and is in a dynamic state of formation and evolution [13, 14]. For those with relatively young formation ages, there may be only gushing and seepage of gas or fluids under the sea floor and associated biological communities but no methane carbonate pavements. In contrast, due to the long-time frame of the formation and

evolution and the influence of tectonic movement, either the gas under the sea floor leaks out or the passage may be blocked by carbonate rocks or gas hydrates, so the gas plume and seepage of the cold seep to diminish and cease and the biological communities gradually die out, leaving only large-scale authigenic carbonate rocks [15–18].

Fluid seepage/leakage associated with a cold seep system is a natural phenomenon that widely exists in the Earth system, especially in marine environments [19–23]. This term refers to the complex network formed by the migration of deep gas-bearing fluids from the deep crustal rocks and sediments to the subsurface strata and seabed through different types of migration pathways, which are able to transport oil and gas and other fluids from the deep strata into the shallow gas hydrate stability zone (GHSZ) where gas hydrates can precipitate and accumulate [13, 24, 25]. In addition, a large amount of gas may even escape into the water column and atmosphere through the submarine leakage pathway, causing changes in the sedimentary environment of the seafloor and the Earth's surface [26, 27]. Therefore, the seepage/leakage associated with cold seep systems is often significant in the fields of energy, ocean and marine ecosystems, and global climate. This unique but widely distributed geological phenomenon has received increasing amounts of attention from researchers and has become a hot topic in related fields.

Unconventional oil and gas, e.g., shale gas, oil sands, coal-bed gas, and gas hydrates, are receiving more and more attention from governments, the oil and gas industry, and petroleum geologists [28–30]. Unlike other unconventional oil and gas resources, gas hydrates are a clean, pollution-free, energy source with high energy intensity and a wide distribution in permafrost and continental slope deep water areas and thus have a huge resource potential [31]. Additionally, as one of the more environmentally friendly energies that may replace traditional carbon resources in the future, gas hydrates have attracted special attention [32, 33]. Many countries and research institutions have invested a great deal of capitals and research effort into geological research and resource evaluation of gas hydrates [34, 35]. In addition, offshore gas hydrate production tests have been successfully implemented in the Nankai Trough and the Shenhu area in the northern South China Sea (SCS), demonstrating the potential and hope for large-scale commercial exploitation of gas hydrates in the near future [36–38].

Cold seeps are commonly found in submarine basins in the continental and shelf regions, such as in the Gulf of Mexico [39, 40], the Blake Sea [12, 41], and the Mediterranean Sea [42, 43]. Moreover, the activities of cold seeps in deep-water areas are closely related to gas hydrate systems [13, 15, 44]. First, the presence of a cold seep, especially the seeping of methane-rich gas, often provides hydrocarbons for the formation and accumulation of gas hydrates. Second, the dissociation of gas hydrates may directly result in the formation of a cold seep system. Finally, the coverage of a cold seep generally indicates the distribution area of the gas hydrates. Therefore, detecting and investigating cold seeps are one of the more effective means of exploring gas hydrates in deep sea areas. A large number of gas hydrate samples

have been recovered from cold seep development areas, e.g., the Gulf of Mexico, the Black Sea, and the SCS [45–53]. In addition, gas hydrates with shallow burial depths and high saturations are usually precipitated in the area of a cold seep system. This phenomenon has been observed in the Nankai Trough, the Gulf of Mexico, and offshore of India where pore-filling and fracture-filling gas hydrates with high saturations have commonly been recovered [54–56]. The existence and dissociation of gas hydrates may cause geological and geochemical processes such as variations in the microtopography, the precipitation of carbonate rocks, methane leakage, the propagation of microbial communities, and even exposed gas hydrate mounds on the seafloor [39, 47, 48, 57–59], which are significant indications of the presence of gas hydrate accumulations. Therefore, the observation and sampling of submarine outcrops have become an important and effective method in the investigation of gas hydrate systems. The identification of microgeomorphological types related to seafloor seepage and the establishment of gas seepage patterns suitable for specific areas can provide a corresponding basis for gas hydrate prospecting. In addition, there are usually a series of microtopographical features in the seabed in gas hydrate occurrence areas, such as slumps, pockmarks, mounds, mud volcanoes, depressions, and platforms [27, 60–64]. Identifying and characterizing these seafloor features not only is of great significance to the discovery and confirmation of the existence of gas hydrates but also provides basic engineering geological information for the exploration and development of gas hydrate resources.

Most of the gas hydrate accumulation areas discovered or presumed based on global exploration have been confirmed or speculated to have a close relationship with the oil and gas migration, seepage systems, and associated submarine cold seep systems in specific areas, such as the Black Sea [24, 65], the Gulf of Mexico [25, 66], the Mediterranean Sea, the Caspian Sea [42, 67], the Hikurangi continental margin offshore of New Zealand [68, 69], and the Nankai Trough [70, 71]. Gas hydrates and conventional petroleum drilling and sampling have also demonstrated the existence of the close relationship between shallow gas hydrates and medium-deep oil and gas reservoirs in most petroliferous basins, mainly because the extensive oil and gas leakage from the deep reservoirs and source kitchens provide sufficient water and hydrocarbon sources for the formation of shallow gas hydrates. This has been demonstrated in the Gulf of Mexico Basin where cold seeps commonly occur and gas hydrates have been recovered from most deep-water oil and gas boreholes [45, 72]. In summary, the hydrocarbon and fluid migration associated with cold seep systems not only plays an important role in the migration and accumulation processes of conventional oil and gas reservoirs but also plays an important role in the gas supply for gas hydrate accumulation.

The northern SCS is a passive continental margin, in which the Neogene tectonic activity was intense, and there are two main structural layers composed of Paleogene rift sequences and Neogene depression sequences. The abundant terrestrial clastic rocks derived from Southern China

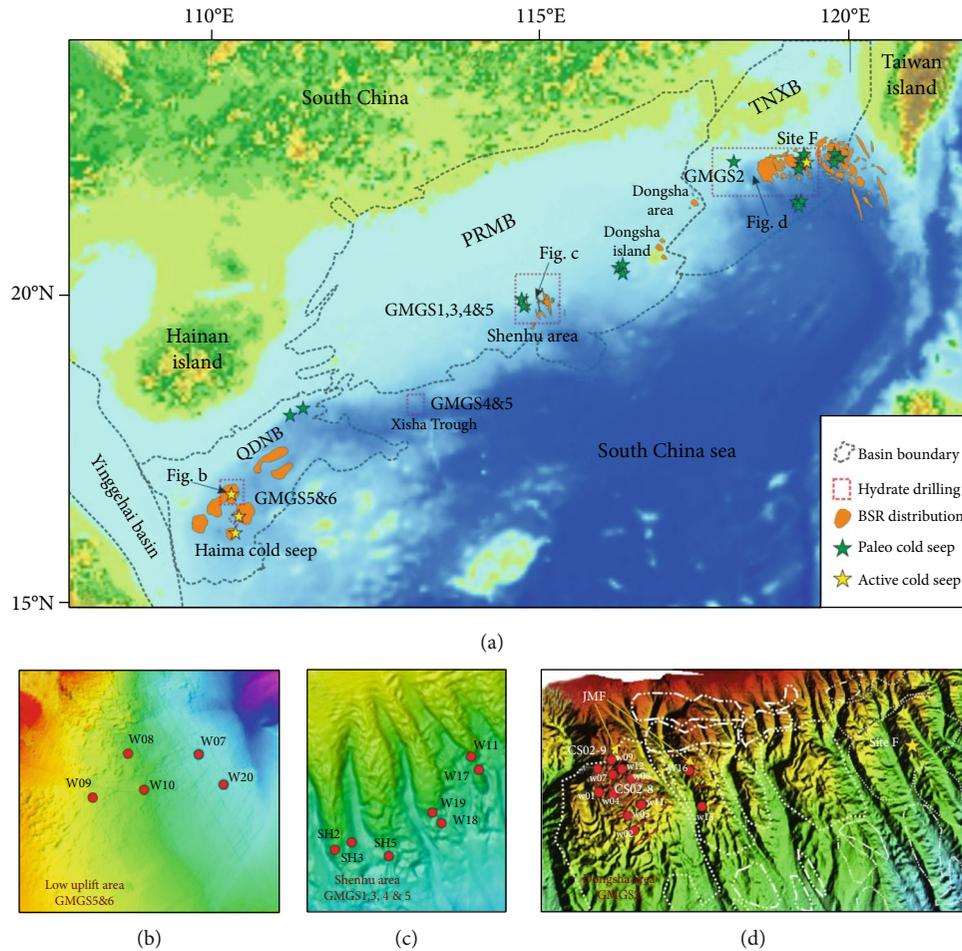


FIGURE 1: (a) Distribution of cold seep systems associated with gas hydrate accumulations in the marginal basins in the northern South China Sea. The distribution of the BSR is from Wang et al. [120], and the distribution of the cold seep system is modified after Feng et al. [6]. (b) Gas hydrate drilling zone and partial sampling sites in the low uplift area of the Qiongdongnan Basin (QDNB) for expeditions GMGS 5 and 6 [120]. (c) Gas hydrate drilling zone and partial gas hydrate drilling/coring sites in the submarine ridges in the Shenhu area in the Pearl River Mouth Basin (PRMB) for expeditions GMGS 1, 3, and 4. (d) Distribution of the gas hydrate drilling sites implemented in expedition GMGS2 and the locations of the Jiulong Methane Reef (JMF) and cold seep Site F (yellow star) in the Taixinan Basin (TXNB). The red circles indicate the location of the gas hydrate drilling/coring sites, and the white dotted lines indicate the distribution of the BSR, modified after Konno et al. [53].

and the Indochina peninsula have resulted in Neogene sediment deposits up to ~10 km thick in the center of several petroliferous basins. The thick stratigraphic deposition and rich hydrocarbon generation and expelling processes of the deep source kitchens have led to active fluid activity, forming a series of NE trending basins that are rich in oil and gas, with widely distributed mud diapirs, mud volcanoes, gas chimneys, and submarine cold seeps [62, 73–78]. Numerous acoustic surveys, including 2D and 3D seismic, subbottom profiles, and multibeam investigations, as well as extensive deep sea diving observations, sampling, and gas hydrate drilling and coring [17, 49–51, 76, 79–82], have been carried out in the northern SCS. The number of cold seeps and associated gas hydrate accumulations being discovered is continuously increasing. Since the discovery of the first cold seep system in the northern SCS in 2004, more than 40 active and paleo-cold seeps have been discovered in the Qiongdongnan Basin, the Xisha area, the Shenhu area, the Dong-

sha area, and the Taixinan Basin [6, 83]. Among them, the cold seep found at Site F offshore of southwest Taiwan in 2013 [17] and the Haima Cold Seep discovered in the deep water Qiongdongnan Basin in 2015 are typical active cold seep systems (Figure 1) [49, 50, 84]. Since 2007, the Guangzhou Marine Geological Survey (GMGS) has carried out six gas hydrate scientific drilling expeditions in the Shenhu area and the Xisha area (GMGS1, GMGS3, and GMGS4), the Dongsha area (GMGS2), and the Qiongdongnan Basin (GMGS5 and GMGS6) in the northern SCS [51, 74, 76, 79, 80, 82, 85–91]. Except for the Xisha area, gas hydrate samples have been recovered in several regions where cold seep systems have been discovered. In addition, the Chinese Academy of Sciences has also found exposed seafloor gas hydrates related to a submarine active cold seep in the Taixinan Basin [47, 48, 53]. These gas hydrate accumulations, which are related to the development and evolution of the cold seep system, are mainly distributed in the vicinity of

the low uplift, submarine ridge, and the channel-levee system and on the seabed above the deep faults, large mud diapirs, and gas chimneys. Therefore, the development and evolution of cold seeps and the precipitation and accumulation of gas hydrates may be closely related to the deep strata and petroleum system. However, at present, most previous studies have focused on the activities of cold seeps and their possible indications for gas hydrate accumulation in shallow and subsurface sediments. Furthermore, there is a lack of research on deep-shallow coupling of submarine cold seeps, associated gas hydrate accumulations, and deep petroleum systems. Therefore, the purposes of this review are (1) to investigate the seafloor geomorphology and geological and geophysical characteristics of the cold seep systems and associated gas hydrate accumulations that have been drilled and sampled in the northern SCS; (2) to discuss the relationship between gas hydrate accumulation and the development and evolution of cold seeps; and (3) to determine the deep-shallow coupling mechanism of cold seep systems, their associated gas hydrate accumulations, and deep petroleum systems. We hope this review will prove helpful in cold seep research and gas hydrate exploration in the SCS and regions with similar geologic settings.

2. Submergence Observations and Sampling in Cold Seep Systems

Based on the comprehensive interpretation and analysis of 2D/3D seismic data, subbottom and multibeam data, and manned deep submersible (HOV) and/or unmanned deep submersible (ROV) observations and sampling, scholars are able to investigate submarine cold seep systems in situ, and our understanding of the cold seep system in the northern SCS has been greatly improved [92–102]. Through submergence observations and sampling, several regions of a cold seep system and its associated gas hydrate accumulations have been discovered (Figure 1(a)). However, active cold seeps have only been discovered in the Qiongdongnan Basin, the Dongsha area, and the Taixinan Basin. The detailed results of the investigation are presented below.

In 2004, China and Germany jointly conducted the SO-177 Expedition in the deep water area near Dongsha on the northern continental slope of the SCS, and for the first time, giant distributions of carbonate rocks formed by the seepage from cold seeps were discovered. These cold seep authigenic carbonates are mainly distributed on two submarine ridges with water depths of 550–650 m and 750–800 m, covering an area of 430 km², which is the largest authigenic carbonate distribution area discovered so far worldwide [92, 93]. This area was named the Jiulong Methane Reef (Figure 1(d)). Chemoautotrophs were found in the cracks of the carbonate crust of the Jiulong Methane Reef. According to isotopic dating, the carbonate crust in this area began to form about 46,700 years ago and is still releasing methane gas. In 2012, an underwater unmanned vehicle (ROV) was used by the GMGS to obtain chimney-like cold seep carbonate rock samples from the Dongsha area (Figure 2(a)). These seabed outcrop observations and sample collection revealed that there are a large number of cold seep vents in this area.

Among them, the paleo-cold seep area is mainly the remains of the paleo-chemical reef, and there are almost no bivalve and bacterial mats. However, benthic organisms and bacterial mats were observed in the current active area of the cold seep. In addition, the Jiulong Methane Reef was later confirmed to be a gas hydrate accumulation area through deep gas hydrate drilling by the GMGS in 2013 [85, 86, 94]. Fracture-filling gas hydrates with various occurrences, including massive, laminated, nodular, and veins, were confirmed to be distributed in multiple layers, and the obtained carbonate rocks indicate that multiple periods of cold seep activity have occurred, which were related to the dissociation of gas hydrates in the past [52]. In the same year, the Jiaolong HOV conducted its first scientific research voyage in the active cold seep area of the Jiaolong-1 site (Site F) to the southwest of Taiwan, and the intermittent activity of this cold seep system was initially identified [17] (Figure 2(b)).

In the deep waters of the Taixinan Basin offshore of southwest Taiwan, scholars not only collected authigenic carbonate rock samples that record cold seep activity from five sites on the Gaoping Slope [95] but also discovered very active cold seeps on the seafloor of the Formosa Ridge (Figure 2(c)). Through ROV in situ observations, living organisms such as methane mats, mussels, and white crabs living in hydrothermal vents were observed on the seabed [96]. In addition, wide distributions of bottom simulating reflectors (BSRs) have been confirmed offshore of southwest Taiwan via high-resolution seismic data [97] (Figure 1(a)). The increasing geological, geophysical, and geochemical findings indicate that the gas hydrate resources in the Taixinan Basin have a great potential [98–100].

In 2015, the GMGS conducted ROV surveys of five sites in the deep water Qiongdongnan Basin using the self-developed Haima ROV, 4500-meter unmanned submersible vehicle in order to collect image evidence related to gas hydrates and to obtain samples of carbonate rocks and chemoautotrophs related to active submarine cold seeps. Three sampling sites were chosen in the southern low uplift area of the Qiongdongnan Basin, and push core and piston core samples containing massive gas hydrates were obtained at two of these three sites [49, 50, 77]. A giant active cold seep, which was named the Haima Cold Seep, with water depths of 1350–1430 m and an area of ~618 km², was discovered for the first time in the northwestern area of the SCS (Figure 2(d)). Piston coring was used to directly sample the shallow surface sediments in the Haima Cold Seep. Physical samples of massive gas hydrates were successfully obtained from depths of 7.95–8.20 m and 4.95–5.10 m below the sea floor at the two sampling sites with water depths of 1381 m and 1405 m [49, 50]. New understandings of the gas hydrate accumulation mechanism and the development of the seafloor ecosystem related to cold seeps in the SCS were gained, promoting subsequent cold seep investigation and gas hydrate drilling site optimization in the Qiongdongnan Basin [77]. In 2018, the QH-ROV-2018 voyage led by the GMGS carried out continuous observations in the Haima Cold Seep area and the northeastern deep-water area of the Qiongdongnan Basin in order to determine whether there are geomorphic phenomena related to submarine leakage

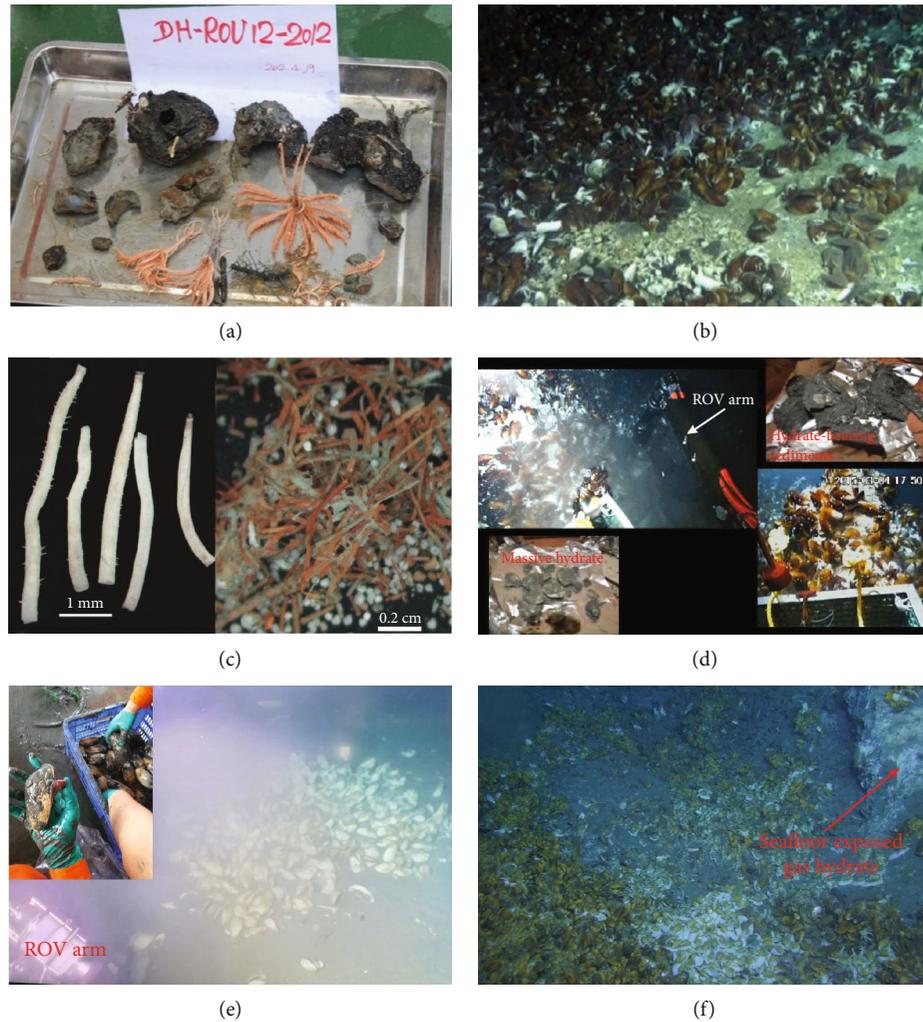


FIGURE 2: (a) Chimney-like carbonate crusts recovered from the Julong Methane Reef. (b) Microbial communities found at the active cold seep in Site F [17]. (c) Tube worms and specimens obtained from the Gaoping slope in the Taixinan Basin [95]. (d) Carbonates, bivalve communities, and massive gas hydrates recovered from the Haima Cold Seep. (e) Bivalve communities discovered by the Haima ROV in a new cold seep developed in the GMGS5 gas hydrate drilling zone, northeastern the Haima Cold Seep. (f) Microbial communities, carbonate crust, and gas hydrates exposed on the seafloor, discovered in a new cold seep near the Haima Cold Seep.

and microbial communities related to active cold seeps in the selected survey sites (Figure 2(e)). These observations provided more direct information for the exploration, drilling, and sampling of gas hydrates in a later GMGS5 expedition [51, 101]. Through Haima ROV observations, seabed microbial communities were found in two of the main investigation sites. Using the sampling manipulator arm, sampling grab, and shallow drilling rig of the Haima ROV, bivalve organisms were collected, and carbonate rock samples were obtained via push coring [102]. In the same year, the Haima ROV of the GMGS and the Deep Sea Warrior HOV of the Sanya Deep Sea Research Institute conducted a joint deep-diving investigation in the Haima Cold Seep and its eastern area, which provided abundant precious information and data for further research of the cold seep and for evaluation of its gas hydrate resource potential [84] (Figure 2(f)).

3. Geophysical Indications of Cold Seeps and Gas Hydrates

There are usually a series of microtopographic features in the seabed in gas hydrate accumulation areas, such as slumps, pockmarks, mounds, mud volcanoes, depressions, platforms, and cold seeps. Identifying and characterizing these seafloor features not only is of great significance to the discovery and confirmation of gas hydrate systems but also can provide basic engineering geological data for the exploitation of the gas hydrate resources. Through a comprehensive study of the seismic, subbottom profile, and ROV image data for the target area for cold seep and gas hydrate exploration in the northern SCS, a variety of seismic and acoustic reflection features related to fluid seepage/leakage were recognized, including gas plumes, seabed mounds, pockmarks, acoustic blanking, and acoustic turbidity [47–50,

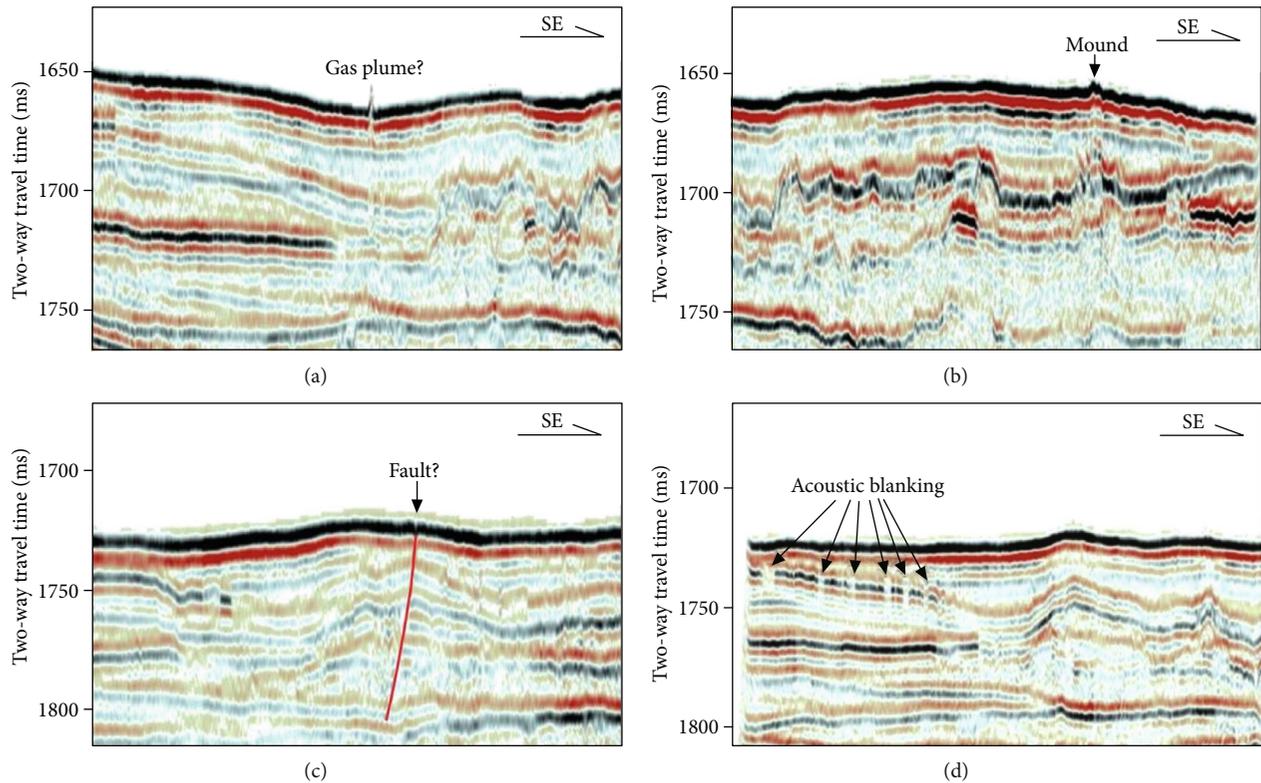


FIGURE 3: Seismic profiles showing the seabed microgeomorphological features related to gas seepage in the deep-water Qiongdongnan Basin.

62, 64, 77, 103–106]. In addition, microbial communities, bacterial mats, methane biochemical reefs, and carbonate crusts associated with cold seeps have been found in the Qiongdongnan Basin, the Xisha Trough, the Dongsha area, and the Taixinan Basin in the northern SCS (Figure 2) [17, 47–49, 52, 101, 107, 108]. These seafloor seepage phenomena, especially the discovery of gas plumes, suggest that the gas source in the study area is sufficient. The appearance of a large number of bivalves and bacterial mats is often a sign of a cold seep with active methane seepage, while carbonate crusts may indicate the cessation of methane seepage. These signs also indicate that gas hydrates are likely to accumulate in the cold seep area.

Based on the fine interpretation of a 3D seismic profile in the Qiongdongnan Basin, many geophysical anomalies closely related to gas seepage were identified in the deep-water area (Figure 3). In addition, a large number of seabed pockmarks, mounds, and acoustic blanking reflections, which are indicative of the presence of free gas and gas hydrates, were identified on the subbottom profile (Figure 4). The reflection of the acoustic blanking reflects the gas-bearing strata, which is consistent with the local structural high, indicating the accumulation characteristics of the gas at the high point. Small mounds, i.e., tens to hundreds of meters in diameter, were interpreted on top of the acoustic blanking, indicating the accumulation and seepage of gas from depth. The presence of large flourishing biological communities often indicates the presence of current methane seepage, which is likely related to the dissociation

of the gas hydrates precipitated in the shallow strata. The commonly occurring microtopography of the seabed and the acoustic anomalies suggest that there is distinct hydrocarbon leakage, indicating a good prospect for gas hydrate occurrence in the deep water Qiongdongnan Basin.

On the seismic profiles, the most favorable indicator of gas hydrate occurrence is a BSR, which exhibits a high amplitude mimic of the seafloor with a negative reflection polarity and usually crosscuts the surrounding strata. A BSR is generally recognized in the overlapping strata of the low uplift and slope area of the Qiongdongnan Basin, but it is usually difficult to identify in deep sags due to the parallel deposition of the Quaternary strata (Figure 5). A BSR was clearly identified by the seismic anomalies resulting from the gas migration along the low uplift, with a fuzzy seismic reflection zone below the BSR (Figures 5(a) and 5(d)). The gas seepage pathways extending from the BSR to the seafloor were identified on the high-precision 3D seismic profile, which shows the “pulled-up” features of the events on both sides of the vertical pathways (Figure 5(a)). Pulled-up reflectors are usually caused by the presence of a material that is harder than the surrounding strata since the hard material can cause velocity anomalies [53, 109]. The gas hydrate drilling and sampling conducted during GMGS expeditions 5 and 6 have demonstrated that hard materials, including gas hydrates and authigenic carbonate rocks precipitated within the seepage/migration pathways, correspond to the pulled-up features [76]. The piston coring and push coring also recovered gas hydrates and authigenic carbonate rocks from

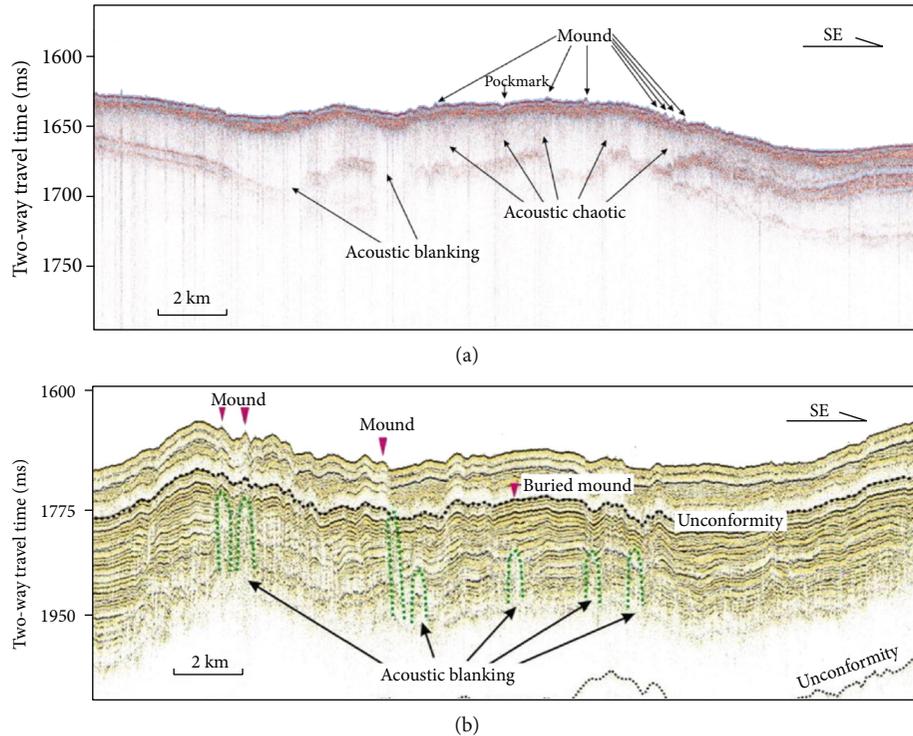


FIGURE 4: (a) Subbottom profile and (b) seismic profile showing the seabed microgeomorphological features related to gas seepage in the deep-water Qiongdongnan Basin.

the subsurface sediments in the seepage sites in the Qiongdongnan Basin [49, 50]. The fracture-filling gas hydrate drilling and ROV diving site optimization in the Qiongdongnan Basin mostly targeted the seepage pathways with pulled-up features, and it was found that these seismic anomalies represent the occurrence of gas hydrates associated with carbonate rocks [50, 51, 76]. In addition, the seepage pathways usually correspond to cold seep vents on the seafloor, and this phenomenon has been commonly observed in the Haima Cold Seep (Figures 5(a)–5(c)). Anomalies with various amplitudes that are associated with seafloor seepage were observed on the seismic profiles, mainly including high amplitude bright spots associated with BSRs and large areas of blanking or chaotic reflection zones caused by mud diapirs and/or gas chimney, which promote gas migration and accumulation in the deep strata [77]. Gas hydrates and seeps have also been discovered in the shallow strata over mud diapirs and gas chimneys in the Shenhu area, the Dongsha area, and the Qiongdongnan Basin in the northern SCS.

Gas chimneys with chaotic seismic reflections and pull-down features indicating the migration and accumulation of hydrocarbons, as well as BSRs and associated seismic anomalies indicating the accumulation of gas hydrates, have been identified by interpreting the quasi-3D seismic data for the Dongsha area [53, 76, 85, 86]. In addition, a large number of slumps and listric faults have been identified in the shallow strata. The above characteristics indicate the occurrence of gas seepage in the area and the possibility of the development of various submarine microgeomorphologies associated with gas seepage. A large number of blanking zones was found on the subbottom profiles [105, 106]

(Figure 6(a)), indicating that there may be mud diapirs with a high strata pressure or faults conducive to fluid migration in this area. Based on the analysis of the subbottom profile, the acoustic blanking reflection anomalies indicating the presence of mud volcanos and gas accumulation in the shallow sediments have been observed (Figure 6(b)), and it was found that there are many pockmarks that may have been caused by submarine gas leakage [105, 106] (Figures 6(c)–6(h)). When the deep gas-bearing fluid is blocked by the shallow strata and cannot reach the seafloor, the upper strata will be deformed due to the overpressure and will form dome features in the seafloor, which is also one of the macroscopic manifestations of gas seepage below the sea floor (Figure 6(d)). Gas chimneys are commonly observed on the seismic profile in the Dongsha area [110]. In general, the internal reflections of these gas chimneys are disordered and chaotic or are characterized by acoustic blanking. In addition, the continuous reflection events on both flanks are suddenly interrupted and terminate at the edge of the chimney zone. The pulled-down phenomenon of the event can be commonly seen on the top of the gas chimney, and the bright spots on both sides and the top of the upper part are distinct (Figure 6). The low amplitude, pulled-down features are mostly caused by the low-velocity anomaly created by the gas charging. The bright spot is the enhanced reflection formed by the accumulation of free gas. Most of the gas chimneys extend from the Miocene strata to the Quaternary strata. In plan view, the gas chimneys are mainly located in the eastern and western ridges, indicating that the local structural high controls the hydrocarbon migration and accumulation (Figure 7). Vertically, the large gas

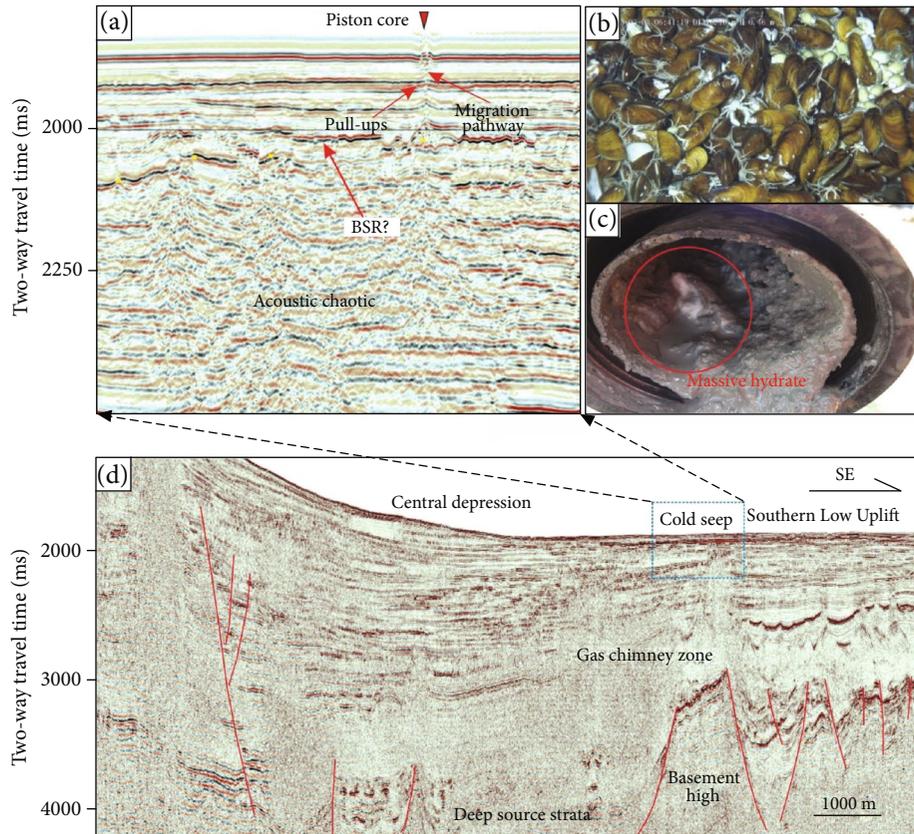


FIGURE 5: (a) Seismic profile showing the hydrocarbon migration and seepage features in the Haima Cold Seep. (b) Microbial communities and (c) massive gas hydrates recovered from the Haima Cold Seep (modified from [49]). (d) Seismic profile showing the deep structural features below the Haima Cold Seep in the southern low uplift area of the Qiongdongnan Basin. Large range of acoustic fuzzy zones resulting from gas chimneys can be observed on the seismic profile, indicating the upward migration of deep hydrocarbons, which supply materials for gas seepage and gas hydrate accumulation.

chimneys in the drilling area almost terminate below the BSR, indicating that the gas-bearing fluid is enriched at the base of the gas hydrate stability zone (BGHSZ) during the upward migration, and gas hydrates precipitated under the appropriate temperature and pressure conditions [53, 62, 97, 111]. That is, the gas chimneys control the hydrocarbon migration and gas hydrate formation and accumulation in the Dongsha area. In addition, mud diapirs and mud volcanoes have also developed in the deep-water Taixinan Basin offshore southwest of Taiwan. They exhibit a large range of vertical acoustic blanking and chaotic reflections on the seismic profile, and domes and cone-shaped structures are commonly observed in the seafloor. Continuous BSRs have been interpreted in the vicinity of the mud volcanos, mud diapirs, and gas chimneys, indicating the presence of gas hydrates (Figure 8).

Similar to the Dongsha area, the seismic profiles in the Shenhu area show that there are a large range of fuzzy reflection zones formed by diapirs and/or gas chimneys, which have a columnar or mushroom appearance in the vertical direction [53, 76, 85, 86]. Bright spots were observed on the top and edges of the gas chimneys, indicating the presence of active gas-bearing fluids in the Shenhu area. BSRs are widely distributed in the study area, and very high amplitude BSRs occur in the upper parts of the gas chimneys [103,

104]. A set of amorphous enhanced reflections that are oblique to the BSR usually occur immediately above the BSR. Drilling has confirmed that these enhanced reflections indicate the presence of gas hydrates [80, 112] (Figure 9). With the acquisition and interpretation of high-resolution 3D seismic data, large-scale listric faults connecting the deep source kitchens, medium-deep petroleum reservoirs, and shallow GHSZ were observed in the gas hydrate accumulation zone [103, 104, 113]. In addition, it has been found that the gliding faults in the GHSZ are partially connected with the BSRs and extend upward to the seabed, constituting possible pathways by which gas can enter the GHSZ or escape and leak into the water column after gas hydrate dissociation due to slope failure. Although no active gas seepage or cold seeps have been confirmed at present, paleo-cold seep activity has been confirmed through geochemical testing of sediments recovered from the Shenhu area [114–116].

4. Distribution of Gas Hydrate Accumulations Associated with Cold Seeps

The seafloor observations and sampling, seismic exploration, and drilling of gas hydrates have confirmed that almost all of the regions containing cold seeps in the northern SCS are favorable regions for gas hydrate accumulation [48–51, 76,

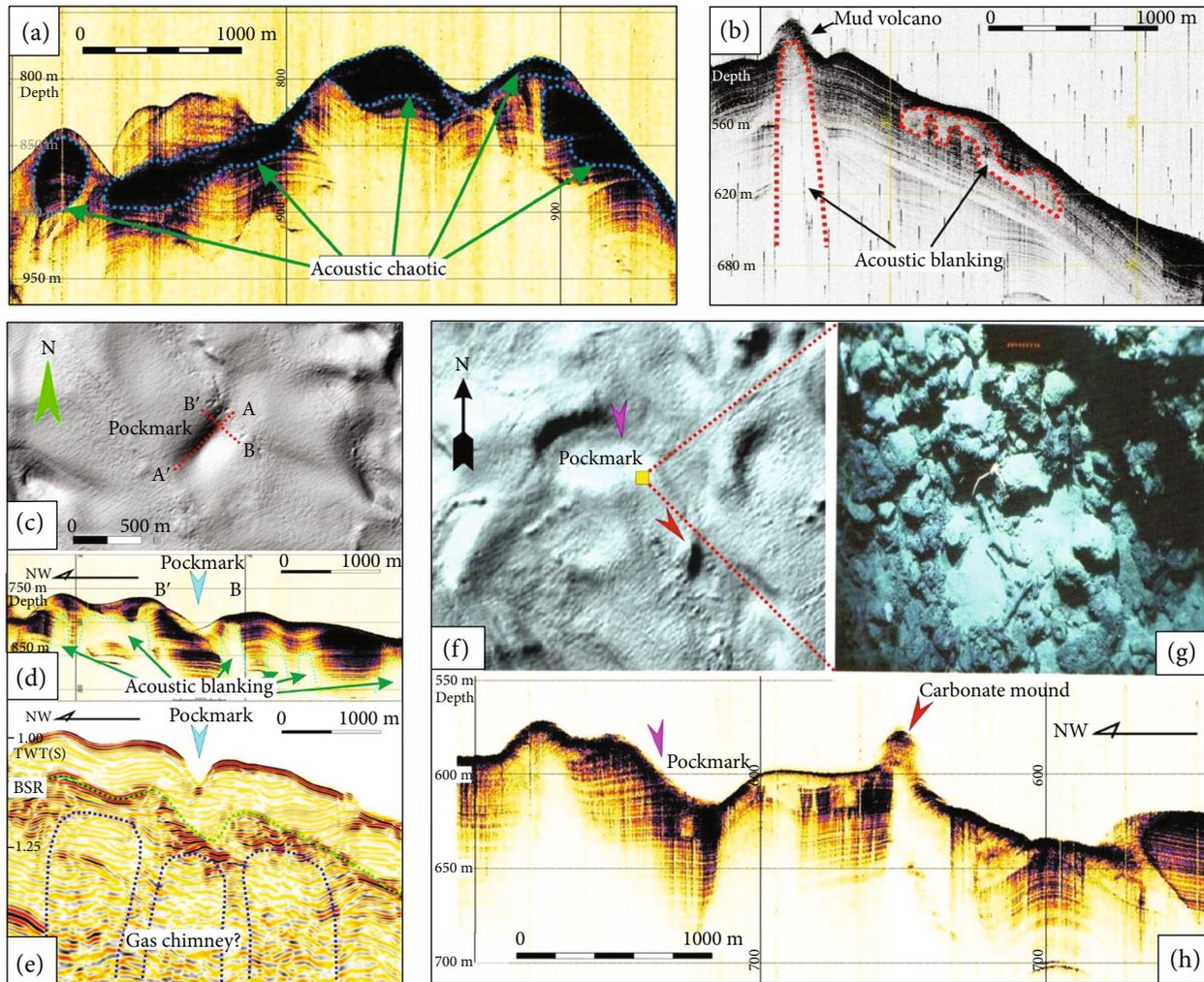


FIGURE 6: Subbottom profile and seismic profile showing the microgeomorphological features associated with the cold seep and gas hydrate accumulation in the Dongsha area (modified from Shang et al. [105] and Shang et al. [106]). The pockmarks are closely related to the gas-bearing fluid migration from the deep strata, which are indicated by the acoustic blanking and the chaotic features on the subbottom and seismic profiles. The submarine mound with authigenic carbonate pavement resulted from gas hydrate dissociation and the seepage of hydrocarbons.

86, 117, 118]. Gas hydrate accumulation related to cold seep activity has been found in the development area of the paleo-cold seep in the Shenhu area, the development area of the active cold seep in the deep water Qiongdongnan Basin, and the development areas of the paleo-cold seep and active cold seep in the Dongsha area and the Taixinan Basin.

The GMGS1, GMGS3, and GMGS4 gas hydrate drilling expeditions and many geological surveys have been conducted in the Shenhu area, more than 60 boreholes have been drilled, and pore-filling gas hydrates with high saturations have been recovered from ~160 to 240 mbsf through pressure coring at sites SH2, SH3, SH7, W11, W17, W18, and W19 (Figure 1(c)) [74, 80, 87–90, 112]. In addition, authigenic carbonates associated with the strong ancient anaerobic oxidation of methane (AOM) have also been recovered, indicating a paleo-cold seep in the Shenhu area [118, 119]. Massive shallow sediments have been recovered

from piston coring sites 08CF7 and Site4B in the Shenhu gas hydrate accumulation area. Studies have shown that the collected authigenic carbonate minerals have a cold-seep origin, and their formation indicates seafloor methane seepage and methane anaerobic oxidation (AOM) [115, 116]. In addition, the discovery of cold seep carbonate rocks reflects the continuous seepage of methane-rich fluids in this area, and it is speculated that the decomposition of gas hydrates is an important reason for the cold seep activity in this area [114]. Although the presence of gas hydrates has not been confirmed through drilling at these piston coring sites, the extensive distribution of BSRs and a large number of boreholes drilled in other parts of the Shenhu area have confirmed the widespread presence of pore-filling gas hydrates [80, 87–91, 112]. The geological and geophysical data also show that there are mud diapirs, gas chimneys, and associated fault systems in this area [103, 104, 113]. In

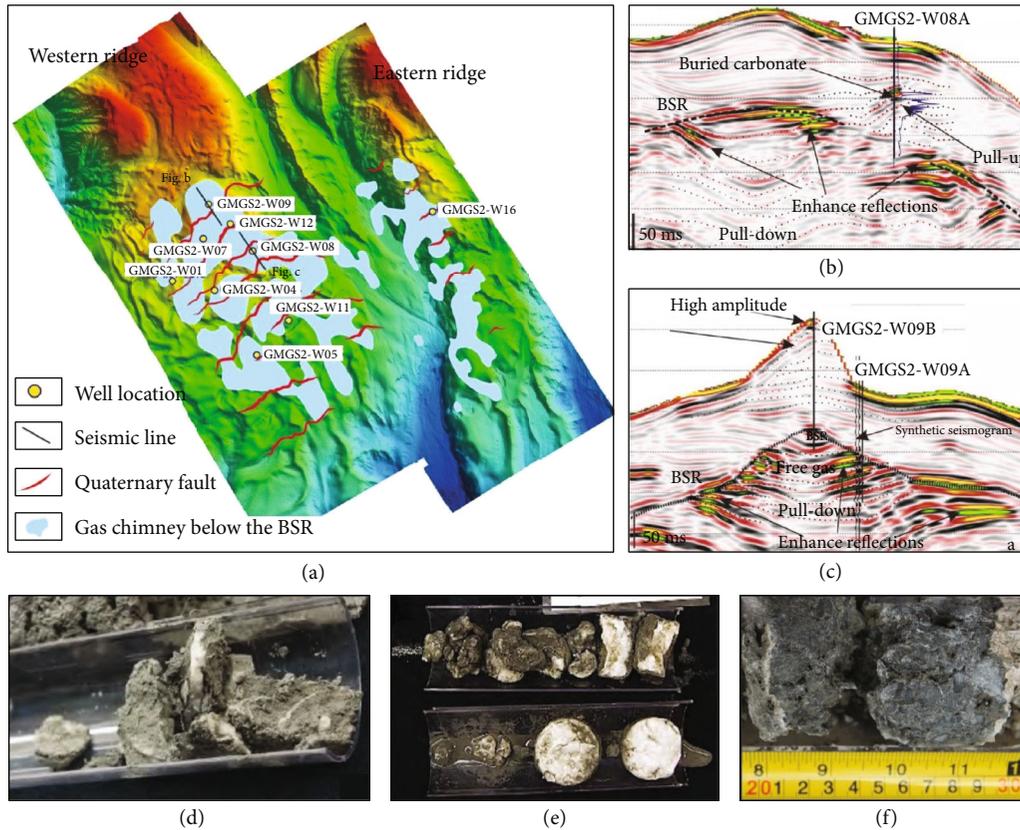


FIGURE 7: (a) Distribution of gas chimneys, faults, and gas hydrate drilling sites for the GMGS2 expedition drilling zone in the Dongsha area. The locations of the drilling sites are from Konno et al. [53] and Zhang et al. [85]. (b) and (c) Seismic profiles showing the gas hydrate accumulation features at the drilling and coring sites, where gas hydrates and buried carbonates were confirmed via logging and pressure coring (modified from Konno et al. [53]). (d) and (e) Massive gas hydrate samples collected from site GMGS2-W08 [133]. (f) Carbonate rock recovered from site GMGS2-W08 (modified from Zhong et al. [94]).

plan view, the distribution of the BSR correlates well with the distribution of the gas chimneys (Figure 9), indicating that the abundant gas supply contributes to the formation and wide distribution of the gas hydrates. Gas hydrate drilling expeditions GMGS1, 3, and 4 in the Shenhu area have demonstrated that the coring sites in which high saturation gas hydrates (up to 67%) were drilled and obtained are directly coupled with the gas-bearing migration pathways below the BSR [80, 81, 103, 104, 113]. However, the accumulation of high saturation gas hydrates tens of meters thick may have prevented further seepage of the gas below the GHSZ into shallow strata and the seafloor, resulting in almost no seepage under the current geological setting.

The BSR in the deep central sag belt and southern low uplift area in the Qiongdongnan Basin has been interpreted to have a wide distribution, indicating the extensive presence of gas hydrates (Figure 1(a)), and increasing geological surveys and drilling programs have been conducted in this area in recent years [77, 120]. In 2015, massive gas hydrate samples were obtained via piston coring of the shallow surface several meters below the seafloor in the Haima Cold Seep in the southern low uplift [49, 50]. The gas hydrates are mainly distributed near the gas seepage vent, which is closely related to the deep free gas. In the Haima Cold Seep area, a

large number of gas seepage/leakage vents have been observed on the seafloor [84]. The vertical pathways in the seafloor can be directly observed on the seismic profile, and a large amount of gas hydrates has accumulated near these pathways [49, 121]. Gas hydrate drilling expeditions GMGS5 and GMGS6 were conducted after the QH-ROV-2018 ROV survey expedition, and they discovered a new cold seep system in the deep-water Qiongdongnan Basin. Fourteen sites were drilled and logged, and except for one site, all of the sites targeted fracture-filling gas hydrates within cold seep systems. In 2018, deep gas hydrate drilling expedition GMGS5 was carried out in the cold seep area at the top of the vertical seepage pathways formed by large gas chimneys in the eastern Qiongdongnan Basin, where massive, nodular, and layered gas hydrates were recovered via pressure coring of the shallow strata (~200 mbsf) at sites W07, W08, and W09 (Figure 1(b)). The gas hydrates were mainly precipitated in the fractures and small faults within the GHSZ [51, 76, 82]. Such fractures and small faults formed via gas fracturing not only constituted pathways through which the deep gas leaks into the seabed but also provided the space for gas hydrate accumulation. Therefore, the seafloor cold seep vents confirmed by the ROV and the leakage pathways identified on the seismic profile in this area

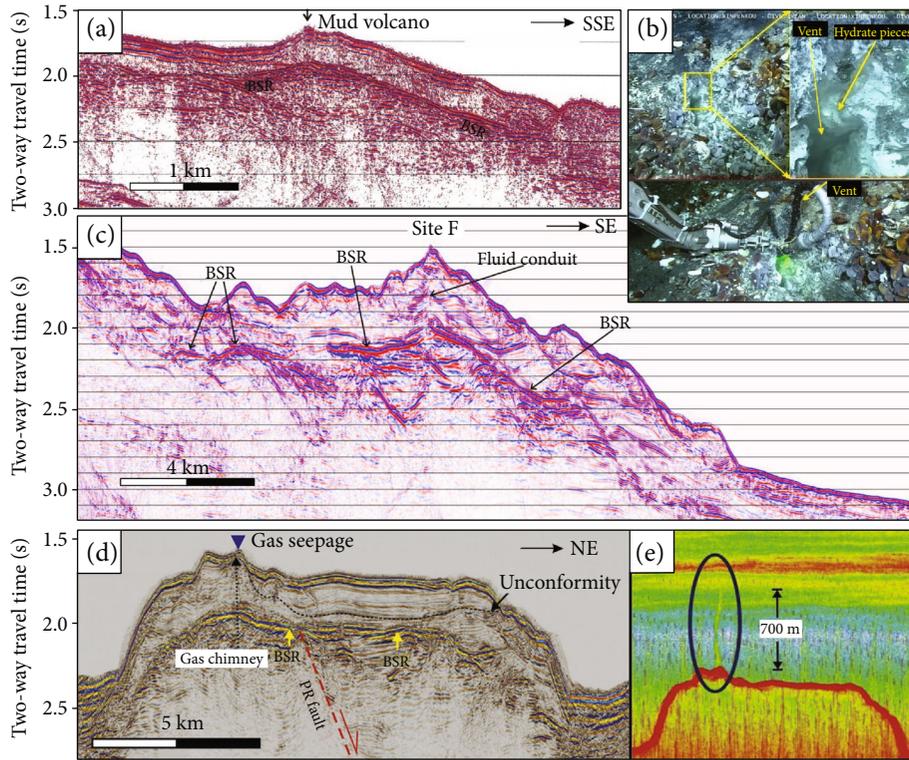


FIGURE 8: (a) Seismic profile showing the BSRs observed in the vicinity of a mud volcano developed offshore of southwestern Taiwan (modified from Liu et al. [97]). (b) Cold seep vents and gas hydrates exposed on the seafloor at site F on the Formosa Ridge [47]. (c) Seismic profile showing the gas seepage and BSRs crossing site F on the Formosa Ridge [124]. (d) Seismic profile showing the gas seepage and BSRs on Pointer Ridge [126]. (e) Gas plume detected on Pointer Ridge [126]. Fault PR is the Pointer Ridge fault.

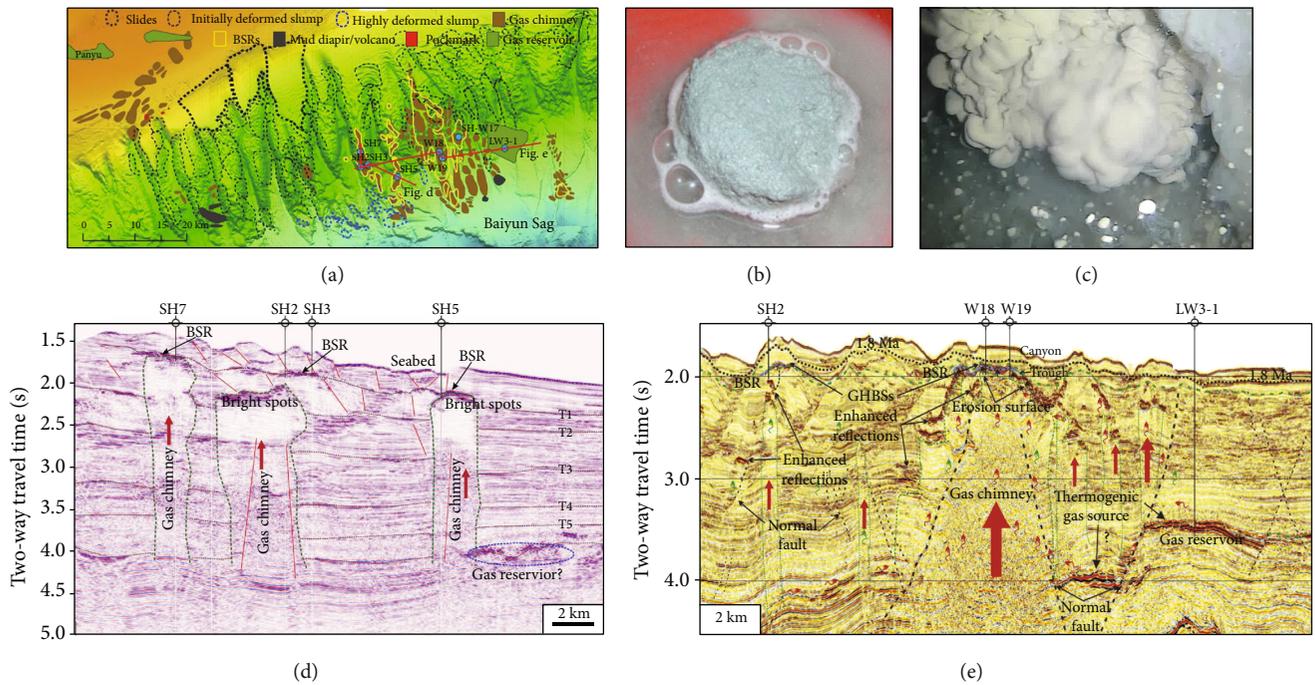


FIGURE 9: (a) Overlapping relationship between the gas chimneys, mud diapirs, mud volcanos, pockmarks, BSRs, and gas reservoir in the Shenhu area (modified from Chen et al. [103]). (b) Dissociation of pore-filling disseminated gas hydrate sample. (c) Gas hydrate floating during deep drilling [81]. (d) and (e) Seismic profiles showing the geophysical reflection characteristics of the gas hydrate drilling sites in the Shenhu area (modified from Jin et al. [138]).

are direct indicators of the occurrence of gas hydrates. In addition, carbonate rocks have been drilled in the seabed of the cold seep area, which are the product of the dissociation of gas hydrates. This shows that the change in the GHSZ was accompanied by a variation in the deep gas flux during the cold seep activity, which resulted in multistage gas hydrate formation and dissociation [82]. In 2019, a new active cold seep system was discovered by the Haima ROV in the eastern area of the Haima Cold Seep in the deep-water Qiongdongnan Basin. Gas hydrates directly exposed on the seabed and accompanied by large gas seeps were observed, indicating that some of the gas hydrates are in the process of dissociation [122].

The seismic interpretation and gas hydrate drilling in the Dongsha area have demonstrated that the distribution of the BSR is well correlated with the distribution of the gas chimneys and NE trending listric faults in plan view. In 2013, the GMGS carried out gas hydrate drilling expedition GMGS2, which targeted the gas hydrate accumulation within the Jiulong Methane Reef in the Dongsha area of the eastern Pearl River Mouth Basin. A total of 23 boreholes were drilled and logged at 13 sites (Figure 1(d)), and a large number of gas hydrate samples with various occurrences, including massive, veins, laminated, nodular, and disseminated, were recovered from multiple gas hydrate-bearing layers within 220 mbsf at sites W05, W07, W08, W09, and W16 [85, 86]. The gas hydrates generally accumulated in the top strata with distinct leakage features in the gas chimney zone (Figures 7(b) and 7(c)). However, the cold seeps in the drilling area are currently inactive, and the seabed is covered by a large area of carbonate pavement and dead microbial communities [53]. Carbonate samples have also been obtained via drilling and coring of the GHSZ, indicating that gas hydrate dissociation accompanied the cold seep in geological history [94]. In addition, an active cold seep with distinct gas seepage and microbial communities has been discovered at Site F southwest of Taiwan, and exposed gas hydrates were found on the seafloor [47, 48, 53]. In plan view, most of the active and inactive cold seeps are located within the distribution area of the BSR (Figure 1). There is also an obvious connection between the gas hydrates and the seafloor seepage, and the gas hydrates accumulated at the top of and/or on both sides of the vertical seepage pathways (Figure 7).

5. Gas Hydrate System

5.1. Gas-Bearing Fluid Migration. Through further studies in recent years, marine petroleum geological research and exploration has confirmed that the deep-water cold seeps, gas hydrate accumulations, and associated seismic anomalies, including BSRs, in the northern SCS are closely related to the gas-bearing fluid migration pathways, which are controlled by the tectonic and sedimentary conditions. A gas hydrate system is genetically related to the focused fluid flow and seepage systems composed of mud diapirs, mud volcanoes, gas chimneys, pipes, and faults in the different regions [49, 53, 76–78, 80, 85, 121, 123, 124].

The drilling sites of expeditions GMGS1, GMGS3, and GMGS4 in the Shenhu area were located on the top of gas

chimneys or mud diapirs, which provided the pathways by which the deep gas-bearing fluids could migrate into the shallow strata and transported the thermogenic gas generated by the Paleogene source rocks and the shallow biogenic gas in the Baiyun Sag to the GHSZ to form gas hydrates [112, 113]. At the drilling sites from which high saturation gas hydrates (~60%) were recovered through pressure coring, such as sites W18 and W19, the gas-bearing fluids migrated upward from the deep Paleogene strata along large-scale mud diapirs and gas chimneys, resulting in large fuzzy seismic zones on the seismic profiles [77]. In plan view, the development range of the diapirs and gas chimney has a very obvious relationship with the distribution of the BSR and the gas hydrate accumulations, which are mainly distributed along the ridge (Figure 9). This demonstrates that the migration pathways of these gas-bearing fluids controlled the gas hydrate accumulation in the Shenhu area [104, 112, 113]. Based on the high-resolution 3D seismic data, it was found that there are also large, deep faults in the Shenhu area that connect the Paleogene source rocks and the shallow GHSZ, which also serve as vertical pathways for fluid migration, especially the thermogenic gas derived from the deep source rocks [104, 112]. Within the GHSZ, due to sediment gliding and/or slope failure, some of the gas hydrate-bearing strata have formed slump faults, which connect the base of the GHSZ with the seafloor, constituting pathways by which the gas entering the GHSZ can move further upward, and some of the gas may leak out of the seafloor after the dissociation of the gas hydrates, producing upward fluid migration. Additionally, the gas-bearing fluids commonly migrate along the preexisting slump faults developed above the BGHSZ, and this fluid escape may in turn cause seafloor collapse and gas hydrate dissociation.

Medium-strong seafloor gas seepage is occurring in the deep-water Qiongdongnan Basin, and a large number of seepage pathways related to gas hydrate formation and accumulation have developed, including mud diapirs, gas chimneys, and faults of variable scales [77, 78]. The distribution of the BSR indicates that the occurrence of the gas hydrates has a direct vertical and planar superposition relationship with these fluid activities [77]. Most significantly, the seismic reflections below the Haima Cold Seep discovered in 2015 are chaotic, creating a large fuzzy zone due to the migration of deep gases into the shallow strata. In addition, several faults and fractures have been identified within the cold seep area, constituting vertical pathways for deep fluid migration and seepage [49, 121]. In 2018 and 2019, massive gas hydrate samples were successfully obtained from a large gas chimney development area in the eastern Qiongdongnan Basin [51, 76, 82]. Seismic interpretation and gas hydrate drilling have demonstrated that this gas hydrate accumulation is closely related to the underlying gas chimneys and deeply buried low uplift. The large faults developed on both sides of the uplift directly connect the deep source kitchens with the vertical gas chimneys and the GHSZ, acting as thermogenic hydrocarbon migration pathways [76, 125].

Since the Late Miocene, Neotectonic activity has occurred in the cold seep and gas hydrate accumulation area

in the Dongsha area. A large number of gas chimneys, mud diapirs, and mud volcanoes, which act as gas-bearing fluid migration pathways, have developed in this area. In particular, clusters of gas chimneys have been identified on the seismic profiles in the GMGS2 gas hydrate drilling area [53, 64]. Another type of hydrocarbon migration pathway in the Dongsha drilling area is faults, which are mainly inherited faults and small active faults on the top and/or flanks of the large gas chimneys, and some of the inherited faults extend downward into the deeply buried basement. The fault development is usually accompanied by gas chimneys or is located directly inside the gas chimneys. These faults have been active since the Late Miocene and are mainly distributed in the western ridge in the drilling area. However, the faults developed in the eastern ridge are less abundant and are smaller in size (Figure 7). Most of the identified faults are subvertical and steeply dipping and developed from the Late Miocene to the Quaternary. Some of these faults even reach the seafloor. Many of the faults developed on the top and/or flanks of the gas chimney are steep in occurrence, small in size, and variable in strike. They often cut the BSR and are associated with enhanced reflections, which are the result of the upward migrated hydrocarbons along the gas chimneys and deep large-scale faults. When the gas flux is sufficient, gas hydrates will form and accumulate in the GHSZ. In addition, the gas hydrate accumulation area corresponds well with the strike of the faults and the extension direction of the gas chimneys (Figure 7) [85]. The gas hydrate drilling and coring sites in the Dongsha area are basically located near the fault at the top of a gas chimney (Figure 7). The successful recovery of fracture-filling gas hydrates indicates that the highly efficient fluid migration system composed of faults and gas chimneys enabled high flux gas-bearing fluid migration and gas hydrate accumulation in the Dongsha area, and it also controlled the formation, accumulation, and distribution of the gas hydrates [110].

In the eastern Dongsha area, the mud volcanoes and mud diapirs near the BSRs are well developed in the Taixinan Basin, indicating that they constitute the pathways for the migration of the deep gas-bearing fluids needed for gas hydrate formation [97]. Gas chimneys, pipe structures, and unconformities, which constitute the pathways by which the gas-bearing fluids migrate into the GHSZ, have also been observed in this area (Figure 8). Based on the seismic profile crossing the Site F (Formosa ridge) cold seep in the Taixinan Basin, the gas chimneys that constitute the migration pathway by which the deep gas leaks into the seabed and forms a cold seep were identified [53, 124]. In addition, mud diapirs and gas chimneys that directly connect the BSR with the seabed and constitute gas migration and leakage pathways were also identified in Pointer Ridge in the Taixinan Basin, forming cold seeps and gas plumes on the seabed [126, 127].

Although the majority of the gas seeps discovered in the northern SCS are related to preexisting pathways composed of faults, gas chimneys, and mud diapirs, there are gas seeps that formed due to the behavior of the gases themselves. Many high-resolution seismic profiles exhibit

indicators of the presence of shallow gas seeps in the Quaternary sediments in the northern SCS, such as pipe structures, bright spots, seabed mounds, and pockmarks (Figures 3 and 4). These subsurface gas seepage/leakage areas are probably caused by gas hydrate dissociation and have nothing to do with the existing deep faults and mud diapirs.

5.2. Types and Origin of the Hydrate Gases. The formation and activity of the cold seep system are accompanied by a large number of substances, including water, gas, various chemicals, and sediments, that leak from the deep strata into the seabed. The migration and supply of gas-bearing fluids provide the material basis for cold seep activity and gas hydrate accumulation. The source and origin of hydrate gas also affect the development scale of cold seeps and gas hydrate reservoirs. The genetic types and potential source rocks of the hydrate gas in the cold seep areas in the northern SCS have been basically characterized through submarine ROV Laser Raman in situ surveys, seabed and subsurface sampling, and geochemical analyses of gas hydrate samples [50, 128–133].

The hydrate gas samples from the shallow surface of the Haima Cold Seep in the Qiongdongnan Basin in the western SCS reveal that methane is the main component, with a concentration of >99.5%, while ethane and propane are typically present in very low concentrations. Most of the carbon isotope values of the methane are lighter than -60‰. The plot of the carbon ($\delta^{13}\text{C}_1$) and hydrogen (δD) isotope values of the methane indicates that the hydrate gas is a mixture of biogenic gas and thermogenic gas, with a bias toward biogenic gas. The carbon isotopes of the ethane range from -25.5‰ to -26.8‰ [50]. The geochemical composition and isotopic characteristics of the hydrate gas indicate that in addition to the shallow biogenic gas supply for gas hydrate formation, deep mature thermogenic gas also contributed to the gas hydrate formation and accumulation [50, 77]. In 2018, the GMGS5 expedition carried out deep drilling in the cold seep area in the eastern Qiongdongnan Basin to recover gas hydrate samples. The geochemical analysis results revealed that the hydrate gas contains up to 20% C_2 + hydrocarbon gas [76, 134]. The $\delta^{13}\text{C}$ and δD values of the methane indicate that the hydrate gas has a mixed genetic origin but predominantly thermogenic gas [125]. In addition, through comparative analysis of the genesis of the shallow hydrate gas and deep conventional natural gas, it has been concluded that there is a close genetic relationship between shallow gas hydrate reservoirs, deep source rocks, and conventional petroleum reservoirs [76]. During the vertical long-distance migration, deep thermogenic gas may undergo microbial degradation and transformation to form secondary microbial methane [125, 134], which together with the shallow biogenic gas supply sufficient material for gas hydrate formation. In general, the cold seep activity and gas hydrate accumulation in the Qiongdongnan Basin are supplied by both biogenic and thermogenic gases (Figure 10).

The samples obtained during gas hydrate drilling in the Shenhu area reveal that the genesis of the hydrate gas varies

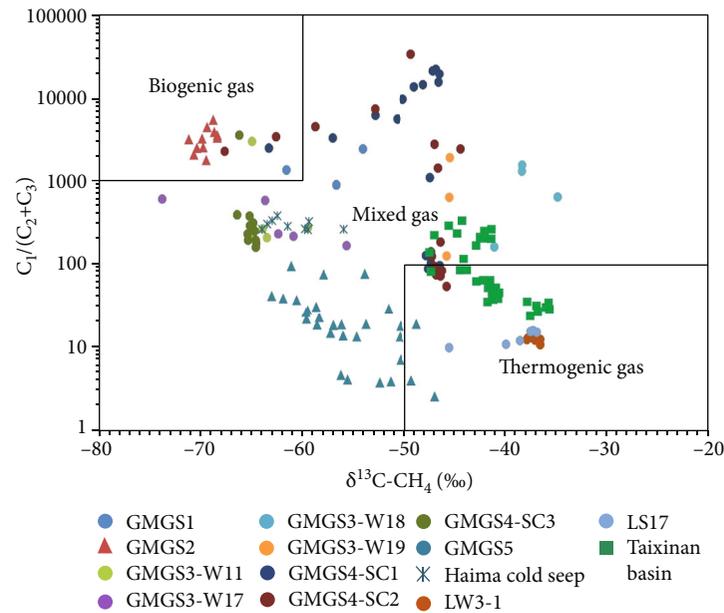


FIGURE 10: Genetic types of the hydrate gases associated with the cold seep system and conventional gas recovered from deep gas reservoirs in the Pearl River Mouth Basin (LW3-1) and the Qiongdongnan Basin (LS17) in the northern South China Sea. The GMGS 1, 2, 3, 4, and 5 hydrate gas data are from Su et al. [74], Zhang et al. [132], Zhang et al. [134], and Sha et al. [141]. The Haima Cold Seep hydrate gas data are from Minshull et al. [50]. The conventional gas data for LW3-1 and LS17 are from Dai et al. [131] and Zhang et al. [155]. The hydrocarbon data for the Taixinan Basin are from Chen et al. [144].

greatly among the different sampling sites. The methane concentrations of the hydrate gas obtained at sites SH2, SH3, and SH7 during the GMGS1 drilling expedition in 2007 were >99.9%, and most of the carbon isotope values of the methane were lighter than -55‰ , indicating biogenic gas [129, 135, 136]. However, it is believed that thermogenic gas also contributed to the gas hydrate accumulation based on analysis of the $\delta^{13}\text{C}_1$ and δD values of the methane and the geological characteristics of the gas hydrate accumulation [74]. According to the results of drilling expedition GMGS3 at sites W11 and W17 in 2015, the methane concentration of the hydrate gas is dominant, and the $\delta^{13}\text{C}_1$ values are basically lighter than -60‰ , indicating microbial gas, while the $\delta^{13}\text{C}_1$ values of the hydrate gas from sites W18 and W19 are greater than -50‰ , suggesting thermogenic gas [132]. The geochemical analysis of the hydrate gas collected from the first gas hydrate production site in the Shenhu area reveals that the hydrate gas is a mixture of biogenic and thermogenic gases [137]. In general, the hydrate gas in the Shenhu area is sourced by a mixture of biogenic gas and thermogenic gas [119, 132], but most of the areas are dominated by biogenic gas (Figure 10). In addition, similar to the Qiongdongnan Basin, the source of the thermogenic gas in the gas hydrate reservoir in the Shenhu area is closely related to the deep gas reservoirs [132, 138]. The genetic type of the thermogenic gas in the confirmed gas hydrate reservoir in the Shenhu area is similar to the gas in the deep conventional reservoirs, such as LW3-1, PY30-1, and PY35-1, which is derived from the hydrocarbon generation supply of the mature Paleogene source rocks in the Baiyun Sag [81, 132, 139, 140].

The methane concentration of the hydrate gas obtained during expedition GMGS2 in the Dongsha area is 96.5–99.8%, and no C_2+ hydrocarbons were detected. The $\delta^{13}\text{C}_1$ values of the methane are -68.4‰ to -71.2‰ , and the δD values are -182‰ to -184‰ , suggesting a biogenic origin (Figure 10) [141]. Although the genesis of hydrate gas obtained from several drilling sites indicates that only biogenic gas contributed to the formation of the gas hydrates, some researchers believe that thermogenic gas that is closely related to the deep petroleum reservoirs contributed to the formation of the gas hydrates in the Dongsha area based on analysis of the biomarkers of the organic matter in the gas hydrate-bearing sediments [142, 143]. Nevertheless, the deep petroleum reservoirs in this area have not been confirmed in any commercial borehole. In situ Laser Raman probing and testing in the Taixinan Basin have shown that there is a large amount of methane gas in the gas hydrates accumulated in the shallow cold seep area [47, 48]. In addition, geochemical studies of the hydrocarbons collected from the mud volcano development area show that the $\delta^{13}\text{C}_1$ values range from -103‰ to -35.6‰ , and most of the values are greater than -50‰ , indicating that the hydrate gases are a mixture of biogenic and thermogenic gases, with a bias toward thermogenic gas. This also indicates the contribution of deep mature source rocks to the gas hydrate accumulation [130, 144].

6. Discussion

6.1. Relationship between Gas-Bearing Fluids and the Seafloor Morphology of the Cold Seep. Submarine gas seepage

and/or leakage can greatly change the seabed's topography, forming pockmarks, authigenic carbonate crusts, mounds, domes, mud volcanoes, and other microgeomorphic features [15, 27, 40, 57, 60, 61, 63, 64, 115, 145, 146]. In addition, chemosynthesis-based communities, unique cold seep carbonate minerals, and the occurrence of gas hydrates are frequently encountered in the cold seep development area. Multiple seafloor geomorphic features have been identified in the distribution areas of cold seeps and their associated gas hydrate accumulations in the northern SCS, indicating a strong connection between deep fluid activities and the shaping of the seafloor's topography and geomorphic features.

In area containing cold seeps and gas hydrate accumulations in the Qiongdongnan Basin, biogenic gas and deep thermogenic gas dominated by methane migrated to and gathered in the shallow strata through migration and seepage pathways, including faults, pipes, diapirs, and gas chimneys, and leaked into the seabed to form cold seeps [49, 77, 121, 123, 147–149]. The rapid migration and flow of gas-bearing fluids often produce bubbling plumes or gas flames on the seafloor. Rapid and abrupt fluxes of gas and water create craters and/or pockmarks in the seabed [150]. Smaller pockmarks may be the result of deep fluid seepage, which can be directly observed on seismic profiles. The high-flux gases and/or liquid hydrocarbons in the Haima Cold Seep region cause strong gas plumes hundreds of meters high [77]. The medium flux fluids can form gas hydrates in the subsurface sediments and even form gas hydrate mounds and accompanying chemosynthetic communities [40], which were verified by the ROV observations in 2018 on the seafloor in the gas hydrate drilling area in the eastern part of the Haima Cold Seep [122, 149]. The corresponding relationship between fluid activities with variable fluxes and the seafloor geomorphology have been observed in the deep-water Qiongdongnan Basin, indicating that the intensity of the deep fluid activities in the different regions of the basin is diverse, which is very similar to the cold seep and associated gas hydrate accumulation area in the Gulf of Mexico [57].

Massive pavements and/or patches of carbonate crust on the seafloor have been discovered in the gas hydrate drilling area in the Dongsha area [151, 152]. The formation of these seafloor deposits was related to the paleo-cold seep plumbing systems composed of deep gas chimneys and faults. It has been proposed that the formation of carbonate crusts is related to the large-scale dissociation of gas hydrates in the past ~30–40 ka [93, 151, 152]. The current cold seep fluid activity in the Dongsha gas hydrate drilling area is weak, so it is hard to find active cold seep vents, gas leakage, and biological communities on the seabed. However, in cold seep development areas such as Site F in the central-eastern Taixinan Basin, there are distinct seepage pathways and/or seabed mounds on the seismic profiles, and seabed mounds, mud volcanoes, and pockmarks have been clearly observed through seabed observations [53, 83, 124]. There are abundant microbial communities associated with the cold seep system, and gas leakage and massive gas hydrates have been directly observed on the seafloor, indicating that some of the

gas hydrates are decomposing. In addition, mud diapirs and mud volcanoes are commonly developed in the Taixinan Basin and are usually accompanied by strong gas-bearing fluid activity. A large amount of deep gas migrated into the GHSZ through the mud diapirs and precipitated as gas hydrates. These phenomena indicate that the gas-bearing fluid activity in the Dongsha area is weak-medium at present, but it may have been very vigorous for a long time during the past hundreds of thousands of years. In contrast, the gas-bearing fluid activity in the Taixinan Basin is relatively intense at present.

Biogenic gas generated by microorganisms in the shallow strata can only be diffused in situ or in unconsolidated porous sediments within a limited distance, and it can be enriched in the GHSZ under high pressure and low temperature conditions through low flux seepage and diffusion with a to form gas hydrates [153]. In general, the intensity of the active gas-bearing fluids in the diffusion-dominated areas is extremely weak. Drilling and coring have demonstrated that a large amount of microbial gas is generated in situ and is diffused within a short distance to form widely distributed pore-filling gas hydrates in the Shenhu area [87–91]. Although geophysical and geochemical evidence has demonstrated the contribution of thermogenic gas to the formation and accumulation of the gas hydrates in the Shenhu area [74, 132, 138, 154], the gas supply was limited and may be restricted to the area in the vicinity of gas field LW3-1. In general, due to the weak gas-bearing fluid flux, seafloor seepage is seldom observed, and there are hardly any seafloor cold seep system and associated seafloor authigenic carbonate pavements. Additionally, evidence of a paleo-cold seep has been found in the Shenhu area, indicating that the dissociation of gas hydrates and gas seepage occurred in the past.

Based on the above discussion of the seafloor morphology, the associated gas-bearing fluid activity in the cold seep system, and the associated gas hydrate systems confirmed in the Qiongdongnan Basin, the Shenhu area, the Dongsha area, and the Taixinan Basin, it is reasonable to conclude that the seafloor morphology of cold seeps is controlled by the intensity of the active deep gas-bearing fluids in the northern slope of the SCS. Therefore, observing and studying the submarine geomorphology and the development characteristics of the gas-bearing fluid and associated microbial communities in a cold seep system are of great significance to gas hydrate exploration.

6.2. Deep-Shallow Coupling of Gas Hydrates, Cold Seeps, and Deep Petroleum Reservoirs. From the above analysis of the origin of cold seep-associated hydrate gas, it was found that hydrate-bound gas, especially thermogenic gas, has a very close genetic relationship with the deep source rocks and/or petroleum reservoirs. The seismic profiles crossing cold seeps and gas hydrate accumulation areas also show that deep gas-bearing fluids can be transported through multiple pathways into the GHSZ to precipitate as gas hydrates or to leak out onto the seabed and form cold seeps.

The cold seeps in the Qiongdongnan Basin are developed on the top of the strata overlying the low uplift of the

pre-Paleogene basement, which resulted from intense magmatic intrusion [84, 123, 138]. In addition, the faults and fractures formed by the uplift of structures led to the generation of active plumping systems [84, 123] connected to the deep Paleogene source rocks, transporting deep hydrocarbons upward and resulting in the formation of gas chimneys and associated fluid leakage under the overpressure. Plenty of thermogenic and biogenic gas migrated along these gas chimneys into the GHSZ to form fracture-filling gas hydrates in mass transport deposits (MTDs) [76]. Analysis of the gas hydrate petroleum system revealed the close relationship between the shallow gas hydrates and deep conventional petroleum reservoirs. As can be seen from the seismic profiles (Figure 5), the deep central channel sand gas reservoir and the deep source strata have a direct vertical coupling relationship with the distribution of the gas hydrates. The geochemical characteristics of the gas hydrates drilled during expedition GMGS5 indicate that the thermogenic gas in the shallow gas hydrate accumulation area is consistent with the origin of the natural gas in deep reservoirs such as LS17-2, LS22-1, and Y8-1 [125, 155, 156]. Therefore, it is believed that the shallow gas hydrates are homologous with the deep gas reservoirs. The gas-bearing fluid provided by the development and evolution of the source rocks in the deep depression, the migration system composed of the low uplift caused by the tectonic activity, the associated faults, and the gas chimneys jointly controlled the formation of the submarine cold seep system and the gas hydrate accumulation in the Qiongdongnan Basin.

The hydrate gas recovered from several coring sites in the Shenhu area was supplied by thermogenic gas, which is also related to the deep oil and gas reservoirs [132, 138]. The large-scale mud diapirs and gas chimneys originate in the Paleogene strata and act as migration pathways connecting the thermogenic gas kitchens with the GHSZ (Figure 9). The elements of the gas hydrate system are consistent in all of the gas hydrate accumulation reservoirs in the Shenhu area [104, 132, 138]. In addition, the higher the gas hydrate saturation, the better the spatial matching relationship between the gas source, migration pathways, and GHSZ [80, 104]. Acoustic chaotic reflections resulting from mud diapirs and gas chimneys have been identified below the BSR at sites W18 and W19 in the GMGS3 drilling area where gas hydrates with >60% saturation have been recovered. Upward gas-bearing fluid migration and charging controlled the formation of the high saturation gas hydrates [80, 81, 104]. Additionally, high angle faults have been identified immediately below the high saturation gas hydrate reservoirs (BSRs) in sites W11 and W17 [80]. In the deeper area, there is a deep-shallow coupling relationship between the shallow gas hydrates and the deep petroleum reservoirs, with gas chimneys, mud diapirs, and high angle faults directly connecting the deep reservoirs and the shallow GHSZ [104]. For example, the deep thermogenic gas in gas field LW3-1 was transported to the GHSZ by large faults and mud diapirs to form high saturation gas hydrates [138], which has been confirmed by the isotopic compositions of the hydrate gas and conventional gas in the Baiyun Sag [119, 132, 138]. Therefore, the effective coupling of the deep gas source, the

migration pathways, and the GHSZ promoted the migration and accumulation of high saturation gas hydrates in the Shenhu area.

There are cold seeps with variable active intensities and associated gas hydrate accumulation in the Dongsha area. However, the direct evidence obtained from geochemical analysis of the available recovered hydrate gas samples indicates that the hydrate gas is microbial in origin and has nothing to do with the deep source rocks. In addition, thus far, no commercial conventional oil and gas reservoirs have been discovered in the Dongsha area, making it more difficult to prove the relationship between the shallow gas hydrates and the deep petroleum reservoirs. However, recent basin modeling and geochemical studies have shown that deep source rocks and possible oil and gas reservoirs may contribute to the formation of shallow gas hydrates [142, 143]. In addition, thermogenic gas associated with mud volcanos has been detected in the nearby central-eastern Taixinan Basin, suggesting a potential coupling relationship between the shallow gas hydrates and the potential deep petroleum reservoir [130, 144].

Based on the above discussion, it is believed that there is a deep-shallow coupling relationship between the cold seep system, the gas hydrate reservoir, and the deep petroleum reservoirs in the northern slope of the SCS. It is reasonable to conclude that the deep petroleum system and the gas-bearing fluid migration system jointly controlled the development of the submarine cold seep system and the enrichment of the gas hydrate accumulation in the northern SCS.

6.3. Significance for Gas Hydrate Exploration. The geological, geophysical, and occurrence characteristics of cold seep systems and gas hydrate systems in different regions of the northern SCS can be characterized by analyzing the geological and geomorphological characteristics of the cold seep systems based on submarine image data obtained from submarine observations, deep penetrating seismic and sub-bottom profiles, and gas hydrate drilling and coring information. Both diffusive type (pore filling dominated) and leakage type (fracture filling dominated) gas hydrates associated with cold seep systems have been confirmed and recovered in the northern slope of the SCS [49, 51, 53, 76, 80, 81, 85–91, 112, 120]. The gas hydrates in the Shenhu area are diffusive type with a pore-filling occurrence, and they are mainly accumulated in sediments dominated by clayey silt. The gas hydrates in the Qiongdongnan Basin are mainly leakage type and are mainly present as fracture-filling hydrates, while both fracture-filling and pore-filling gas hydrates have been discovered in the same drilling site at different depths in the Dongsha area. Therefore, there are significant differences in the accumulation and occurrence characteristics of the gas hydrates in different regions. Gas hydrate drillings in the Shenhu area, the Dongsha area, and the Qiongdongnan Basin have revealed that the gas hydrate-bearing sediments are mainly clayey silt and silty clay with little difference in lithology. From the perspective of the differences in the development of the cold seep systems, the intensity of the activity of the gas-bearing fluids, the types of gas migration pathways, and the deep-shallow

coupling relationship between the GHSZ and the deeply buried source rocks may jointly control the differences in the gas hydrate occurrences and accumulation types.

The methane in the pore filling gas hydrates was mainly derived from the strata in the vicinity of the GHSZ. The methane flux was limited, and it is normally in the dissolved state in the pore water [157]. The gas migrated over a short distance through the pores, fractures, and faults. When the concentration of methane dissolved in the pore water exceeds the thermodynamic equilibrium saturation solubility of the two-phase water-hydrate system, the dissolved methane precipitates to form gas hydrates immediately at the BGHSZ [158]. However, hardly any cold seeps have been observed in the areas with abundant pore-filling gas hydrates due to the relatively low gas-bearing fluid flux and impermeable gas hydrate-bearing sediments. The drilling results have shown that there is a good correspondence between the distribution of the gas hydrates and the BSR in the Shenhu area. The BSR is mainly distributed in the submarine ridge and at the position where the ridge dips into the deep sea plain [80, 103, 104]. In addition, the distribution of the BSR has a good spatial overlap relationship with the development and distribution areas of the mud diapirs and gas chimneys, indicating that the gas migration conditions may have controlled the distribution of the gas hydrates. However, although the mud diapirs and gas chimneys extend upward from the deep strata, they may have had a limited effect on the migration of the deep thermogenic gas, and only a small part of the thermogenic gas may have migrated to the GHSZ to precipitate as gas hydrates. Therefore, the slow diffusion of the shallow biogenic gas and the limited migration of thermogenic gas derived from the deep strata controlled the pore filling gas hydrates in the Shenhu area.

The leakage type gas hydrates supplied by both biogenic gas and thermogenic gas recovered or speculated in the Qiongdongnan Basin, the Dongsha area, and the central-eastern Taxinan Basin in the northern SCS indicate that medium-high flux methane migration to the seafloor via free gas leakage and/or seepage along faults, gas chimneys, pipes, mud volcanoes, and mud diapirs controlled the hydrocarbon migration and gas hydrate accumulation. When gas seeped and/or leaked out to the seabed due to a high fluid flux, it is likely to result in geomorphological features such as pockmarks and mounds, and cold seep microbial communities develop. Additionally, through the metabolic processes of autotrophic biota, authigenic carbonate rocks are prone to precipitate. Therefore, the seafloor microtopography and outcrops of carbonate rocks in cold seep systems in deep water areas are important indicators of the occurrence of leakage type gas hydrates. The leakage type gas hydrate enrichment areas with a similar origin discovered by drilling around the world include the North Cascadia Hydrate Ridge in the western continental margin of the United States [26, 151], the Gulf of Mexico [159, 160], the K-G Basin offshore of India [161], and the Ulleung Basin in South Korea [109, 162]. Therefore, based on comprehensive analysis of seismic, subbottom, and multibeam data, analysis of the tectonic activities and investigation of the geological and geomorphic characteristics of the seabed in areas where gas-bearing fluid

migration system has developed are the main approaches to exploring active cold seeps and leakage type gas hydrate systems [163–167].

Generally, the cold seep systems and their associated gas hydrate accumulations in the northern slope of the SCS have been found to have a close relationship with the deep source rocks and/or petroleum reservoirs through multiple gas-bearing fluid migration pathways. It is also reasonable to propose that conventional oil and gas and gas hydrates are paragenetic in petroliferous basins. Therefore, conventional oil and gas reservoirs and favorable hydrocarbon accumulation areas are also favorable gas hydrate exploration targets. The discovery of a cold seep system is also an important indicator of gas hydrate occurrence in the SCS, and more in situ geophysical investigations and deep drilling are needed. We look forward to the significant discovery of gas hydrate-associated cold seep systems and the advancement of our knowledge of them in the SCS.

7. Conclusions

- (1) Cold seep systems with diverse microgeomorphologies and geological and geochemical features are related to the intensity of the activity of gas-bearing fluids in the northern South China Sea. The activity of the cold seeps in the Qiongdongnan Basin is medium-strong, with abundant gas seepage, plumes, and microbial communities. The active cold seeps in the Dongsha area and the Taxinan Basin are similar to those in the Qiongdongnan Basin, exhibiting a medium-high fluid flux. In addition, the widespread authigenic carbonate pavements in some paleo-cold seeps are related to the dissociation of gas hydrates and cessation of seepage. The fluid activity in the Shenhu area is weak at present, and there is almost no submarine leakage
- (2) The cold seep systems in the northern South China Sea are generally connected with the deep strata through a variety of gas-bearing fluid migration pathways, including mud diapirs, mud volcanoes, gas chimneys, and faults. They are a precondition for the formation of a cold seep, and they also control the differential development of cold seep systems
- (3) The activity of the cold seeps in the northern South China Sea is closely related to the formation and accumulation of gas hydrates, which are usually precipitated in cold seep vents and vertical fluid migration pathways, indicating that the formation of gas hydrates is controlled by the activity of gas-bearing fluid
- (4) The hydrocarbons recovered from cold seep systems and their associated gas hydrate accumulations in the northern South China Sea are generally mixtures of biogenic gas and thermogenic gas, which are related to the deep mature source rocks and petroleum reservoirs

- (5) In the northern South China Sea, there is a deep-shallow coupling relationship between the cold seep systems, gas hydrate accumulations, and deep petroleum reservoirs. It is necessary to pay more attention to the favorable accumulation areas of conventional oil and gas with abnormal gas-bearing fluid activity in the later stages of the exploration of active cold seeps and their associated gas hydrates in the South China Sea

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (no. 42076054, no. 41806071, and no. 41806074), the Guangdong Basic and Applied Basic Research Foundation (no. 2019B030302004), Key Special Project for Introduced Talents Team of Southern Marine Science and Engineering Guangdong Laboratory (Guangzhou) (no. GML2019ZD0102), and the China National Hydrate Project (no. DD20190217, no. DD20190218). The authors wish to thank those that contributed to the success of the China National Gas Hydrate Program Expeditions 1, 2, 3, 4, 5, and 6.

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