# **Chapter 4 Gas Hydrates at Seeps**



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**Abstract** Gas hydrates have been the focus of intensive research during recent decades due to the recognition of their high relevance to future fossil energy, submarine geohazards, and global carbon and climate changes. Cold seep-related gas hydrate systems have been found in both passive and active margins worldwide. A wealth of data, including seismic imaging, borehole logging, seafloor surveys, and coring, suggest that seep-related gas hydrates are present in the western Taixinan Basin and the Qiongdongnan Basin of the northern South China Sea (SCS). Here, we provide an overview of the current understanding of seep-related gas hydrate systems in the northern SCS and underscore the need for more systematic work to uncover the factors governing the interplay of hydrate dynamics and gas seepage and to quantitatively assess the temporal and spatial variability of gas hydrate and cold seep systems.

# **4.1 Introduction**

Gas hydrates are ice-like structures that are composed of hydrogen-bonded water molecules forming a lattice of cages that encapsulate molecules of natural gases, mainly methane (Sloan [1998](#page-10-0)). Large quantities of gas hydrates are stored in the submarine continental margin sediments where the conditions required for gas hydrate formation, including high pressure, low temperature, and sufficient supply of methane, are met. Recent decades have evidenced an increasing interest in gas hydrates because they are considered to represent a potential source of fossil energy

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that could exceed the energy content of all other known fossil fuels combined (Holder et al. [1984](#page-10-1); Collett [2002](#page-9-0)). Additionally, as submarine gas hydrates sequester large amounts of carbon, they are thought to form a great capacitor that could regulate the Earth's climate (Dickens [1999;](#page-9-1) Archer et al. [2009](#page-9-2); Ruppel and Kessler [2017](#page-10-2)). Furthermore, if large-scale gas hydrate dissociation occurs in response to environmental changes, slope failure could be triggered, and the released methane could aggravate ocean deoxygenation and acidification (Sultan et al. [2004](#page-11-0); Biastoch et al. [2011\)](#page-9-3).

Gas hydrate provinces associated with methane seep systems are found in both passive and active margins worldwide (Holbrook et al. [2002;](#page-10-3) Tryon et al. [2002;](#page-11-1) Torres et al. [2004](#page-11-2); Boswell et al. [2012;](#page-9-4) Luo et al. [2016](#page-10-4)). Intensive methane supply in the form of advective methane-bearing fluids or methane bubbles from depth can lead to the formation of massive gas hydrates in the gas hydrate stability zone (GHSZ). As a result, seep-related gas hydrates are usually characterized by shallow burial depths and high saturation. Extensive investigation and exploration of gas hydrates in the South China Sea (SCS) have been taking place since 1999. Seep-related shallow gas hydrate systems were suggested to occur in the western Taixinan Basin and the Qingdongnan Basin, northern SCS, based on geophysical imaging and seafloor observations (Fig. [4.1\)](#page-2-0) (Sha et al. [2015;](#page-10-5) Wang et al. [2018;](#page-11-3) Geng et al. [2021\)](#page-9-5). Indeed, seafloor coring and drilling has confirmed their occurrence in both areas (Zhang et al. [2015;](#page-11-4) Bohrmann et al. [2019](#page-9-6); Hu et al. [2019;](#page-10-6) Wei et al. [2019](#page-11-5); Meng et al. [2021;](#page-10-7) Ren et al. [2022](#page-10-8)). The western Taixinan Basin is located in the northeastern rifted passive SCS margin and is close to the active continental margin where the SCS oceanic crust subducted eastward beneath the Philippine Sea Plate. Horsts and grabens formed across the margin during the period of rifting (Late Cretaceous–late middle Miocene) and were subsequently buried by marine deposits (Clift and Lin [2001;](#page-9-7) Zhu et al. [2009\)](#page-11-6). Due to the general extensional tectonic environment, two groups of normal faults striking NW–WNW and NNE–ENE are well developed in the western Taixinan Basin (Zhang et al. [2015](#page-11-4)). This region is characterized by the widespread presence of NNW–SSE trending topographic ridges due to canyon incision. During the R/V SONNE 177 cruise in 2007, a widely distributed seep carbonate crust called Jiulong Methane Reef was found in the northern part of the western Taixinan Basin, indicating extensive paleo-methane seep events in this area (Suess et al. [2005;](#page-11-7) Han et al. [2008\)](#page-9-8). The Qiongdongnan Basin, located on the northwestern continental slope of the SCS, contains a Cenozoic sedimentary succession of up to 12 km in thickness and is suggested to have great hydrocarbon and gas hydrate potential (Zhu et al. [2009](#page-11-6); Shi et al. [2013](#page-10-9); Zhang et al. [2019](#page-11-8)). A number of mega-pockmarks related to submarine fluid flow together with indicators of shallow gas hydrate occurrence have been reported in the western Qiongdongnan Basin (Sun et al. [2011;](#page-11-9) Luo et al. [2014](#page-10-10)). In 2015, "Haima cold seeps" were discovered in the southern Qiongdongnan Basin during ROV surveys launched by the Guangzhou Marine Geological Survey. Living chemosynthesis-based communities (e.g., mussels, tubeworms and clams), gas ebullition, seep carbonates, and massive gas hydrates were observed and recovered from the "Haima cold seeps" (Liang et al. [2017b\)](#page-10-11).

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**Fig. 4.1** Distribution of cold seep-related gas hydrate systems in the northern South China Sea

<span id="page-2-0"></span>The primary goal of this chapter is to provide an overview of the current understanding of shallow gas hydrate systems associated with methane seeps in the western Taixinan Basin and the Qingdongnan Basin, northern SCS by synthesizing the scientific results obtained in the past decade.

# **4.2 Characteristics of the Shallow Gas Hydrates**

### *4.2.1 Gas Hydrate Textures*

Gas hydrate samples in marine sediments can be described in a variety of ways, namely, as massive layers, lenses, veins, nodules, and disseminated grains, based on their macroscopic appearance. In most cases, shallow gas hydrates exhibit different textures from their deeply buried counterparts. Two drilling campaigns have been launched in the western Taixinan Basin: the GMGS2 expedition in 2013 and the SO266 MeBo200 drilling cruise in 2018. During the GMGS2 expedition, 13 sites were drilled, with 10 sites logged while drilling (LWD), along the crest of two seafloor ridges at water depths ranging between 664 and 1420 m (Yang et al. [2014](#page-11-10)). The primary objective of the GMGS2 expedition was to determine the nature, distribution, and saturation of gas hydrates in this basin. At sites GMGS2-W08, GMGS2-W09, and

GMGS2-W16, shallow gas hydrates with saturations up to  $\sim$  50% of pore space were observed (Sha et al. [2015](#page-10-5)). Gas hydrates in the form of veins and nodules were first encountered  $\sim$  8–10 m below the seafloor (mbsf) at these three sites (Fig. [4.2\)](#page-4-0), where LWD data showed both resistivity and p-wave velocity values (Zhang et al. [2015](#page-11-4); Wang et al. [2018\)](#page-11-3). To the east of the drilled ridges during GMGS2, the active seep area, Site F, or Formosa Ridge, was drilled during SO266. Pore water chloride profiles, borehole logging, and IR imaging data all showed clear indications of hydrate-bearing layers in the upper 15–42 mbsf (Bohrmann et al. [2019](#page-9-6)). The shallow gas hydrates mainly exhibit tubular and nodular appearances. These types of hydrate textures observed during both drilling campaigns are likely indicative of substantial methane supply from the deep subsurface, which is also demonstrated by the observations of seep carbonates accompanied by chemosynthetic bivalve shells both in the retrieved cores and on the seafloor (Chen et al. [2016;](#page-9-9) Wang et al. [2018;](#page-11-3) Bohrmann et al. [2019](#page-9-6)).

Shallow gas hydrates (~4 mbsf) in the Qiongdongnan Basin were recovered for the first time from the "Haima cold seeps" in 2015 (Liang et al. [2017b](#page-10-11)), although negative pore water Cl and positive  $\delta^{18}$ O anomalies, which implied gas hydrate dissociation during core recovery, were previously reported in a pockmark located in the western Qiongdongnan Basin (Luo et al. [2014\)](#page-10-10). The recovered gas hydrate samples appeared in the form of small nodules and thin veins (Liang et al. [2017b;](#page-10-11) Hu et al. [2019\)](#page-10-6). During the GMGS5 expedition in 2018, four sites (W01, W07, W08 and W09) were drilled in the eastern Qiongdongnan Basin, with the aim of exploring the distribution, abundance, and formation mechanism of the massive gas hydrates associated with seeps. Drilled sites W07, W08, and W09 contained both abundant gas hydrates and authigenic carbonate in the sediments from the upper ~6–23 mbsf. Large hydrate veins with various dip angles and thicknesses, hydrate nodules of different sizes, degassing, and soupy sediments caused by gas hydrate dissociation have been observed in shallow sediments (Liang et al. [2019](#page-10-12); Wei et al. [2019\)](#page-11-5). Similar to the drilling results from the western Taixinan Basin, seep carbonate crust and typical seep-related bivalve fragments were also found in the recovered cores and on the seafloor in close proximity to the drilling sites in the eastern Qiongdongnan Basin, which likely indicated the potential linkage of shallow massive hydrates and methane seepage (Ye et al. [2019](#page-11-11)). However, the shallow sediments of the eastern Qiongdongnan Basin hold greater amounts of gas hydrate than those in the western Taixinan Basin, implying a more sufficient methane supply from the subsurface in the eastern Qiongdongnan Basin. A subsequent gas hydrate drilling expedition (GMGS6) in the western Qiongdongnan Basin was launched in 2019, with the aim of characterizing the distribution and accumulation of gas hydrates in the sandy sediment layers associated with Quaternary channel-levee facies and mass transport deposits (MTDs) (Meng et al. [2021\)](#page-10-7). Site W01 was drilled at a currently active cold seep, where authigenic carbonate crust and massive gas hydrates were observed on the seafloor (Ren et al. [2022](#page-10-8)). Fracture-filling gas hydrates present as chunks, veins, and nodules first appeared within the MTDs at depths of  $\sim$  5–28 mbsf at Site W01 (Fig. [4.2\)](#page-4-0). Interestingly, the shallow hydratebearing silty sediments were immediately overlain by a thick authigenic carbonate layer as a cap (Meng et al. [2021\)](#page-10-7).



<span id="page-4-0"></span>**Fig. 4.2** Examples of gas hydrate samples retrieved from shallow sediments in the Qiongdongnan Basin (**a** and **b**) and the western Taixinan Basin (**c** and **d**). Seafloor observations of gas seepages and gas hydrates on the seafloor of the GMGS6 drilling area of the Qiongdongnan Basin are also shown (**e** and **f**) (Modified from Zhang et al. [2015](#page-11-4); Wei et al. [2019;](#page-11-5) Meng et al. [2021](#page-10-7) and Ren et al. [2022](#page-10-8)). Reprinted from Marine and Petroleum Geology, 67, Zhang et al. ([2015\)](#page-11-4) Geological features, controlling factors and potential prospects of the gas hydrate occurrence in the east part of the Pearl River Mouth Basin, South China Sea, 356–367, Copyright (2015), with permission from Elsevier; from Marine and Petroleum Geology, 110, Wei et al. ([2019\)](#page-11-5) Characteristics and dynamics of gas hydrate systems in the northwestern South China Sea—Results of the fifth gas hydrate drilling expedition, 287–298, Copyright (2019), with permission from Elsevier; from Deep Sea Research Part I: Oceanographic Research Papers, 177, Meng et al. [\(2021](#page-10-7)) Quaternary deepwater sedimentary characteristics and their relationship with the gas hydrate accumulations in the Qiongdongnan Basin, Northwest South China Sea, 103,628, Copyright (2021), with permission from Elsevier; and from Journal of Petroleum Science and Engineering, 215, Ren et al. [\(2022](#page-10-8)) Sand-rich gas hydrate and shallow gas systems in the Qiongdongnan Basin, northern South China Sea, 110,630, Copyright (2022), with permission from Elsevier

## *4.2.2 Geochemistry of Hydrate-Bound Hydrocarbons*

The sources of light hydrocarbons bound in shallow gas hydrates have been determined for hydrate samples retrieved from both the western Taixinan Basin and the Qiongdongnan Basin. The molecular ratios of hydrocarbon composition  $(C1/(C2+C3))$  and the stable carbon isotopic compositions of methane ( $\delta^{13}$ C–CH<sub>4</sub>) of the shallow gas hydrates retrieved during GMGS2 in the western Taixinan Basin indicate that the hydrate-bound gas is mainly of microbial origin via  $CO<sub>2</sub>$  reduction (Fig. [4.3;](#page-5-0) Liu et al. [2015;](#page-10-13) Liang et al. [2017a](#page-10-14)), which is the prevailing methanogenic pathway in marine sediments. This finding is consistent with Raman spectroscopic and X-ray diffraction results that reveal hydrate crystallization in a typical type I cubic lattice structure I (sI), with dominant occupancy of methane in both large and small cages (Liu et al. [2015\)](#page-10-13). Moreover, preliminary onboard analysis of the compositions of void gas also implies a major contribution of biogenic hydrocarbons at Site F (Bohrmann et al. [2019](#page-9-6)).

In contrast to the biogenic methane that formed the shallow gas hydrates, the hydrate-bound gas in the Qiongdongnan Basin has a mixed biogenic and thermogenic origin based on C1/(C2+C3) and  $\delta^{13}$ C–CH<sub>4</sub> (Fig. [4.3\)](#page-5-0) (Lai et al. [2022](#page-10-15)). Additionally, the detection of C3+hydrocarbons indicates the contribution of thermogenic gas



<span id="page-5-0"></span>**Fig. 4.3** Genetic diagram of  $\delta^{13}C_{CH4}$  versus C1/(C2+ C3) according to Milkov and Etiope [\(2018](#page-10-16)) that is used to identify the hydrocarbon sources of shallow gas hydrates in the South China Sea. CR– CO2 reduction; F—methyl-type fermentation; SM—secondary microbial; EMT—early mature thermogenic gas; OA—oil-associated thermogenic gas; LMT—late mature thermogenic gas; QDNB— Qiongdongnan Basin; TXNB—Taixinan Basin. *Data sources* Liu et al. ([2015\)](#page-10-13); Liang et al. [\(2017a](#page-10-14)); Liang et al. ([2019\)](#page-10-12); Lai et al. ([2021\)](#page-10-17); Wei et al. [\(2021\)](#page-11-12); Lai et al. [\(2022](#page-10-15))

produced in deep subsurface sediments to the shallow gas hydrate system (Ye et al. [2019;](#page-11-11) Lai et al. [2021](#page-10-17)). Furthermore, both sI and sII types of gas hydrates were found at site W08 by means of Raman spectroscopy and X-ray diffraction (Wei et al. [2021\)](#page-11-12). In-situ temperature measurements at sites W07, W08, and W09 yielded linear geothermal gradients of 102–111  $^{\circ}$ C km<sup>-1</sup>, which are substantially higher than those in the background sediments of the Qiongdongnan Basin (65  $\mathrm{C \, km^{-1}}$ ) (Wei et al. [2019](#page-11-5)). The observed high geothermal gradient was explained as the result of vertical migration of deep-rooted, warm fluids, thereby remobilizing thermogenic hydrocarbons toward the seafloor and contributing to the formation of shallow gas hydrates and cold seeps (Ye et al. [2019\)](#page-11-11).

# *4.2.3 Relationship Between Cold Seeps and Shallow Gas Hydrates*

The drilling area of GMGS2 is located in proximity to the deformation front of SW Taiwan. Several NW–SE trending submarine canyons and seafloor ridges are well developed in the shelf edge down to the lower slope. Faults, gas chimneys, and diapiric structures have been identified from seismic imaging in this area (Wang et al. [2018](#page-11-3)). The gas hydrates drilled during GMGS2 in the western Taixinan Basin are inferred to have been fed by a continuous biogenic gas supply from the Pliocene and Pleistocene strata into the GHSZ via mud diapirs, gas chimneys, and deep-rooted faults. Although the hydrate-bound gas was inferred to be sourced from microbe-mediated  $CO<sub>2</sub>$  reduction, secondary microbial methane derived from the bioconversion of thermogenic organic matter (e.g., oil deposits, coalbeds) also contributed to the biogenic gas supply (Gong et al. [2016](#page-9-10)). Shallow gas hydrates tended to accumulate in the topographic highs on the ridges, where overpressure caused by free gas generation in the deep subsurface drove the upward migration of gassy fluids. Focused gas flow that contributes to cold seeps and near-surface gas hydrates can either be transported by gas chimneys formed close to the seafloor or by sub-vertical faults connecting the tops and/or flanks of large gas chimneys in subsurface sediments to the seafloor. These gas conduits can be traced to at least as deep as the base of the GHSZ. Although seafloor observations show generally weak methane seepage intensity in the drilling area of GMGS-2, multiple authigenic carbonate-bearing and bioclastic layers retrieved by the drilled cores suggest large-scale, intense methane seepage events in the past (Sha et al. [2015;](#page-10-5) Wang et al. [2018](#page-11-3)). In addition, authigenic carbonate and gas hydrates are stratigraphically linked, and analysis of the stable carbon and oxygen isotopic compositions of authigenic carbonate points to vigorous methane seepage likely caused by gas hydrate dissociation (Chen et al. [2016](#page-9-9)). Similarly, Site F, which was drilled during SO266, also exhibits a clear subsurface plumbing system that connects deep gas reservoirs with the seafloor and closely links shallow gas hydrates and authigenic carbonate (Bohrmann et al. [2019](#page-9-6)). It is inferred that the accumulation of free gas beneath the base of the GHSZ causes hydraulic fracturing, and the resulting gas

venting occurs through the GHSZ to the seafloor (Kunath et al. [2022](#page-10-18)). Therefore, it is likely that gas hydrate dissociation close to the base of the GHSZ triggered a significant release of methane toward the seafloor, forming authigenic carbonate and gas hydrates in the near-surface sediments of the western Taixinan Basin.

The drilling areas of GMGS5 and GMGS6 are located in the Qiongdongnan Basin, which is one of the major petroliferous basins in the northern SCS. Seismic imaging showed the widespread occurrence of gas chimneys indicated by acoustic blanking and/or chaotic reflections, which were associated with the development of cold seeps in the Qiongdongnan Basin. Large gas chimneys were found to be underlain by pre-Paleogene basement uplift resulting from magmatic intrusion. Faults that were formed during basement uplift could act as effective pathways for transporting deepseated thermogenic hydrocarbons within the Paleogene source rocks and deeply buried gas reservoirs to shallow sediments in the form of gas chimneys (Geng et al. [2021\)](#page-9-5). In addition, a 3D seismic survey revealed that the widely distributed MTDs and channel systems in the Quaternary strata could facilitate hydrocarbon and gas hydrate accumulation (Cheng et al. [2021;](#page-9-11) Meng et al. [2022\)](#page-10-19). Geochemical analysis reveals the same source of hydrate-bound gas as natural gas in deep hydrocarbon reservoirs (Lai et al. [2022\)](#page-10-15). These observations suggest a direct coupling of gas hydrates and deep source rocks and/or petroleum reservoirs, which represents a unique hydrate system in the SCS.

Likewise, the authigenic carbonate collected from the seafloor and the drilled cores of the Qiongdongnan Basin also allows the fingerprinting of methane seepage events caused by gas hydrate dissociation (Liang et al. [2017b](#page-10-11); Wei et al. [2022\)](#page-11-13). Distinct seeprelated seafloor morphologies, e.g., pockmarks and mud volcanoes, are abundant in the western Qiongdongnan Basin and are also postulated to be associated with the waxing and waning of subsurface hydrate reservoirs (Sun et al. [2011;](#page-11-9) Luo et al. [2015;](#page-10-20) Yang et al. [2021](#page-11-14)). The multiple hydrate dissociation-driven methane seepage events imply highly dynamic hydrate systems susceptible to sea-level fluctuation, bottom water temperature change, and sedimentary loading. In addition to vigorous methane bubbling out of the seafloor, some of the seeping methane is currently being converted to gas hydrates in the shallow sediments, as reflected by the observations of positive pore water chloride anomalies within ~10–50 mbsf at Site W08. Therefore, it is likely that the deep petroleum system and gas hydrate reservoirs controlled the development of cold seeps and the accumulation of shallow high-saturation gas hydrates in the Qiongdongnan Basin (Fig. [4.4\)](#page-8-0).

### **4.3 Summary and Perspectives**

Shallow gas hydrates associated with cold seeps have been discovered in the western Taixinan Basin and the Qiongdongnan Basin of the SCS. Logging data and seafloor drilling confirmed the presence of gas hydrates in the upper ~5–30 mbsf. Additionally, seafloor observations showed bare gas hydrates on the seafloor at the active seep sites in the Qiongdongnan Basin. Shallow gas hydrates are generally fracture fillings,



<span id="page-8-0"></span>**Fig. 4.4** Schematic diagram showing the linkage between the hydrates and seeps at the surface and hydrate/hydrocarbon reservoir at depth

with various fabrics, including massive layers, veins, and nodules, which are typical structures of seep-related hydrates. Light hydrocarbons, originating predominantly from microbe-mediated  $CO<sub>2</sub>$  reduction, are trapped in gas hydrates recovered from the western Taixinan Basin. In contrast, light hydrocarbons bound in gas hydrates of the Qiongdongnan Basin are sourced from a mixture of biogenic and thermogenic gas. The upward migration of free gas through the GHSZ is facilitated by faults that connect the deep petroleum reservoir and gas-charged sediment with the seafloor. Periodic release of free gas leads to the precipitation of gas hydrates and the formation of authigenic carbonate in shallow sediments, featuring a seep-related gas hydrate system.

Although significant progress has been made in understanding gas hydrate systems in the SCS, the multi-stage evolution of cold seeps and hydrate dynamics remain

poorly constrained. More in-depth and detailed examination of the relationship between deep-seated petroleum systems and gas hydrate reservoirs and the coupling of deep and shallow hydrate systems are needed. The SCS is bounded by the convergent margin to the east, rifted margin to the north, and transform margin to the west, thereby offering an ideal opportunity to explore and compare seep-related hydrate systems in various tectonic settings. Moreover, as the seep-related hydrate systems in the SCS exist in a unique setting characterized by multiple tectonic regimes, rapid fluctuations in sediment delivery toward the northern continental margin with glacial–interglacial cycles, and variations in both the temperature and strength of bottom currents, the underlying mechanisms of the spatial and temporal variability of hydrate dynamics and gas seepage in the SCS generally remain elusive. The most important key to addressing the abovementioned open questions is to combine longterm monitoring and numerical modeling approaches to quantitatively assess the temporal and spatial variability of gas hydrate and cold seep systems.

**Acknowledgements** The authors are grateful to Prof. Joris Gieskes for his constructive reviews. Tingcang Hu and Weiding Li are thanked for their help with figures.

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