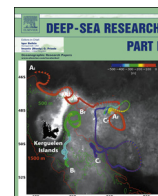




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Formation mechanism of authigenic gypsum in marine methane hydrate settings: Evidence from the northern South China Sea

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ABSTRACT

During the last decade, gypsum has been discovered widely in marine methane hydrate-bearing sediments. However, whether this gypsum is an in-situ authigenic precipitate remains controversial. The GMGS2 expedition carried out in 2013 by the Guangzhou Marine Geological Survey (GMGS) in the northern South China Sea provided an excellent opportunity for investigating the formation of authigenic minerals and, in particular, the relationship between gypsum and methane hydrate. In this contribution, we analyzed the morphology and sulfur isotope composition of gypsum and authigenic pyrite as well as the carbon and oxygen isotopic compositions of authigenic carbonate in a drillcore from Site GMGS2–08. These methane-derived carbonates have characteristic carbon and oxygen isotopic compositions ($\delta^{13}\text{C}$: -57.9‰ to -27.3‰ VPDB; $\delta^{18}\text{O}$: $+1.0\text{‰}$ to $+3.8\text{‰}$ VPDB) related to upward seepage of methane following dissociation of underlying methane hydrates since the Late Pleistocene. Our data suggest that gypsum in the sulfate–methane transition zone (SMTZ) of this core precipitated as in-situ authigenic mineral. Based on its sulfur isotopic composition, the gypsum sulfur is a mixture of sulfate derived from seawater and from partial oxidation of authigenic pyrite. Porewater Ca^{2+} ions for authigenic gypsum were likely generated from carbonate dissolution through acidification produced by oxidation of authigenic pyrite and ion exclusion during methane hydrate formation. This study thus links the formation mechanism of authigenic gypsum with the oxidation of authigenic pyrite and evolution of underlying methane hydrates. These findings suggest that authigenic gypsum may be a useful proxy for recognition of SMTZs and methane hydrate zones in modern and ancient marine methane hydrate geo-systems.

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1. Introduction

Gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) is found mainly in sedimentary and evaporitic environments as an evaporite mineral. However, non-evaporitic occurrences of gypsum have occasionally been reported (Blanchet et al., 2012; Pirlet et al., 2010, 2012). The most common formation mechanisms for non-evaporitic gypsum include the oxidation of sulfides, the action of sulfuric acid solutions on calcium-bearing rocks, and the hydration of anhydrite (Chang et al., 1998). In recent years, gypsum has been frequently observed in marine methane hydrate settings, in which it is hypothesized to

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have formed authigenically (Chen et al., 2007; Kocherla, 2013; Novikova et al., 2015; Pierre et al., 2012, 2014; Sassen et al., 2004; Wang et al., 2004). Although some studies of marine methane hydrate settings have ascribed gypsum formation to oxidation of sedimentary sulfide minerals such as pyrite (FeS_2) (e.g., Pierre et al., 2012, 2014), its authigenic origin has not been adequately demonstrated to date. Furthermore, little consideration has been given to the relationship between gypsum formation and underlying methane hydrate. The present study addresses this gap in our knowledge concerning the formation mechanism of non-evaporitic gypsum and its relationship to co-genetic pyrite in marine methane hydrate geo-systems.

The GMGS2 expedition was carried out in 2013 by the Guangzhou Marine Geological Survey (GMGS) as the second Chinese exploration of gas hydrate-bearing sediments in the northern

South China Sea, during which 13 sites were drilled, with nine sites yielding natural methane hydrates, five of which were sampled for geochemical analysis (GMGS2–05, –07, –08, –09, –16) (Sha et al., 2015; Zhang et al., 2014, 2015). These sediment cores provided an excellent opportunity to study the formation of authigenic minerals in gas hydrate geo-systems and, in particular, the relationship between gypsum, pyrite, and methane hydrates. In this contribution, we analyze the morphologies and sulfur isotopic compositions of authigenic gypsum and pyrite as well as the carbon and oxygen isotopic compositions of authigenic carbonate in the drillcore from Site GMGS2–08, with the aim of determining the formation mechanisms of non-evaporitic authigenic gypsum in marine methane hydrate geo-systems.

2. Geological setting

The investigated area of the GMGS2 expedition lies to the northeast of the Pearl River Mouth Basin (PRMB), where widely distributed bottom simulating reflectors (BSRs) have been observed (Liu et al., 2006; Li et al., 2013; Wang et al., 2006; Zhang et al., 2015). Several expeditions focusing on gas hydrates have been undertaken in this region during the past fifteen years, leading to discovery of substantial quantities of gas hydrate-bearing sediments (Fig. 1) (Han et al., 2008; Yang et al., 2007; Zhang et al., 2007, 2014). In 2004, about 430 km² of methane-derived carbonates, named the “Jiulong Methane Reef”, were discovered in the northeastern South China Sea during the SO-177 cruise (Han et al., 2008). In 2007, China’s first gas hydrate drilling program successfully revealed the presence of thick gas hydrate-bearing sediment layers during the GMGS1 expedition in the Shenhu area of the PRMB (Zhang et al., 2007). In 2013, thirteen sites were drilled during the GMGS2 expedition in the eastern PRMB, of which nine yielded gas hydrate (Sha et al., 2015; Zhang

et al., 2014, 2015). In 2015, the third gas hydrate drilling expedition (GMGS3) yielded a comprehensive set of data and samples that revealed extensive gas hydrate deposits in the Shenhu area of the PRMB (Yang et al., 2007).

Logging-while-drilling (LWD) data indicate the existence of two layers of methane hydrate in the sediments at Site GMGS2–08 (Fig. 2) (Zhang et al., 2014, 2015). The upper methane hydrate horizon is found at depths from 9 to 23 mbsf with a resistivity maximum of 17.5 Ωm at 12 mbsf, whereas the lower methane hydrate horizon is found at depths from 66 to 98 mbsf with a resistivity maximum of nearly 100,000 Ωm at 71 mbsf (Fig. 2). The two zones differ with respect to the density and distribution of methane hydrates, which are present at low density mainly as veins in the upper zone and at high density as massive hydrates in the lower zone (Fig. 2). The resistivity and acoustic velocity logs show increases, and the gamma-ray and density logs decreases, within the methane hydrate intervals (Fig. 2). In addition, authigenic carbonates showing high resistivity and low porosity were discovered at 58–62 mbsf at Site GMGS2–08 (Fig. 2) (Zhang et al., 2014).

3. Materials and methods

Site GMGS2–08 (Fig. 1) was drilled to a depth of 138 m below seafloor (mbsf) using Fugro coring tools at a water depth of 798 m during the GMGS2 expedition from May to September 2013. The coring plan at Site GMGS2–08 was first developed from existing seismic data and was refined with the analysis of logging-while-drilling (LWD) data, as a consequence of which the borehole was not continuous (Zhang et al., 2014). Further details about Site GMGS2–08 and the GMGS2 expedition were provided in Zhang et al. (2014, 2015).

The recovered drillcores were stored at temperatures lower

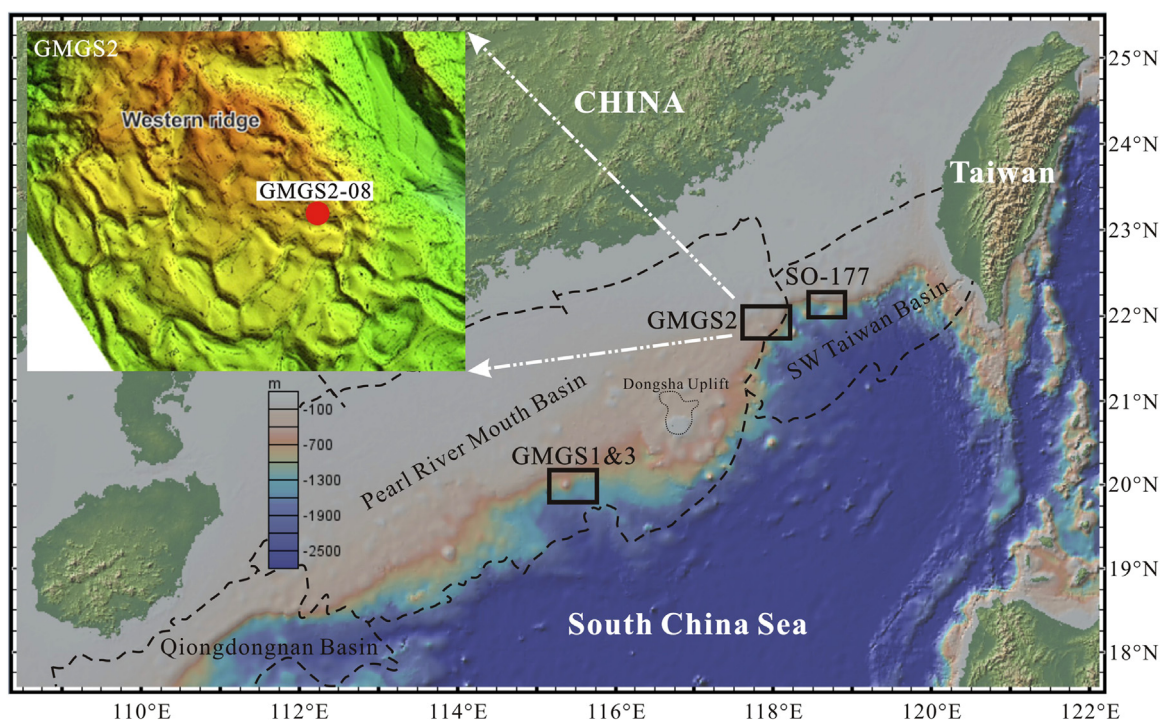


Fig. 1. Location of the study site (red spot) (inset map from Sha et al. (2015)). The SO-177 cruise was carried out in 2004 (Han et al., 2008). The GMGS1 area was drilled in 2007, during China’s first gas hydrate expedition (Zhang et al., 2007), and the GMGS2 area was drilled in 2013, during China’s second gas hydrate expedition (Sha et al., 2015; Zhang et al., 2014, 2015). Recently, the GMGS3 cruise was carried out in the northern South China Sea (Yang et al., 2007). The color schemes for both maps represent seafloor bathymetry; the color legend applies only to the main map. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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