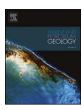
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#### Research paper

## Discussion on the rapid formation mechanism and evolution process of methane hydrate-bearing sediments in Shenhu Area of northern South China Sea



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#### ABSTRACT

Because of free gas migration in sediment pores, the Shenhu seafloor layers in the northern South China Sea is speculated to have experienced some rapid methane hydrate generation and accumulation. In this study, the main hydrate formation is presumed to occur rapidly in the gas-hydrate-liquid three-phase zone. A first-order reaction kinetics mechanism, which controls the hydrate generation, has been coupled into a flow-transportation-reaction model. The methane flux is chosen to be  $0.5 \, \text{mol} \cdot \text{m}^{-2} \cdot \text{a}^{-1}$ , and the scope of the kinetic reaction coefficient ranges from  $10^{-1} \, \text{mol} \cdot \text{m}^{-3} \cdot \text{MPa}^{-1} \cdot \text{a}^{-1}$  to  $10^{2} \, \text{mol} \cdot \text{m}^{-3} \cdot \text{MPa}^{-1} \cdot \text{a}^{-1}$ . Using these basic conditions, two geological scenarios are designed to investigate the hydrate formation within local effective MH distribution area which is 240-150 mbsf. At three moments (800 a, 4 ka and 8 ka) the state of dissolved methane and salt, layer temperature, gas and hydrate saturation, stratum permeability and capillary pressure has been displayed. The effect of kinetic coefficient on gas and hydrate content, and local temperature, has been compared. The phenomenon accompanying transient decomposition at the hydrate formation front has been found in this process. At last, the constant bottom boundary temperature is set to reflect the influence from deep tectonic activity, and the effects have been discussed. The results demonstrate the possibility of a kind of rapid dynamic evolution process and mechanism on hydrate-bearing sediment formation and aggregation. They also present interesting differences between the enclosing and open characteristics of the Shenhu hydrate system.

#### 1. Introduction

The Shenhu hydrate-bearing sediments in the northern South China Sea (SCS) have been speculated to be one type of complex methane hydrate (MH) reservoir that possesses the common characteristics of both diffuse and seepage marine gas hydrate systems (Chen et al., 2006; Fan et al., 2004; Nyuyen, 2012; Tréhu et al., 2006; Wu et al., 2011, 2013; Yang et al., 2015). Geochemistry analysis on sediment pore water indicates that most of the combined methane in gas hydrate primarily originates from microbial activities, while the rest comes from thermogenic gas (Jiao et al., 2015; Lu et al., 2013; Yu et al., 2014). Highresolution seismic profiles (2D/3D) of the Shenhu zone show that hydrate layers are located just above the base of the gas hydrate stability zone (HSZ), and plenty of free gas gathers just below the bottom of the HSZ (Hui et al., 2016; Liu et al., 2012; Wu et al., 2011). Considering that the methane quantity from in-situ sources is insufficient to form

these massive hydrate-bearing layers, where the MH saturation can reach up to  $\sim$  48% in pore, the mixed gas-liquid fluids full of biogenic and thermogenic methane generated from the deep crust have been presumed to permeate up into the HSZ and join the production of MH (Sha et al., 2015).

Besides the appropriate thermodynamic conditions in local sedimentary strata, an abundant methane and water supply is indispensable for breeding a marine hydrate system (MHS) in global continental convergent margins (Paull and Dillon, 2001; Xu, 2004). Theoretically, MH can nucleate and grow in the porous pores under the saturated dissolved methane-bearing single-phase solutions or the free gasbearing two-phase mixed fluids (Sloan and Koh, 2008). In most of actual MHSs, where MHs have massively aggregated within a certain vertical thickness range, local sediment pores generally develop with vast fluxes of dissolved methane and free gas migrated from deeper crust, for example, the MH-bearing reservoirs in Hydrate Ridge (Milkov

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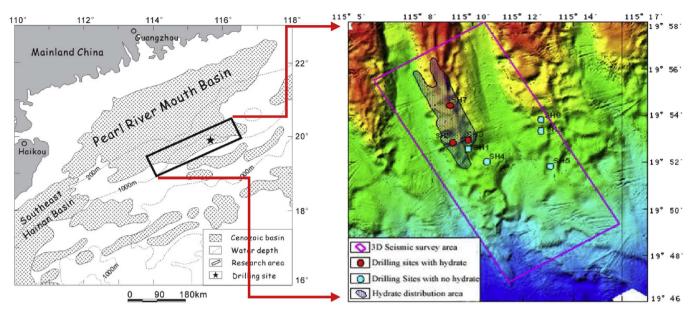


Fig. 1. The geographic map and its topographic profiles of the Shenhu MHS in the northern SCS. In the left square region surrounded by black solid lines three massive MH-bearing layers have been found, indicated by red dots in the right chart (SH2, SH3, SH7). The figure is revised from Su et al. (2012) and the copyright has been authorized from the publisher, Elsevier. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

at al., 2004; Tréhu et al., 2006). These types of active fluids will not only affect the aggregation and occurrence of hydrate-bearing sediments but also influence the formation and distribution of free gas zones and bottom-simulating reflections below the HSZ, and facilitate the morphological growth of bubble plumes and gas chimneys (Cook and Malinverno, 2013; Crutchley et al., 2015; Haacke et al., 2007; Talukder, 2012). The seismic data analysis of similar BSRs, bright reflectors under BSRs, and chimney structures in the Shenhu MHS (Qin et al., 2015; Wang et al., 2014; Wang and, 2017) all suggest that free gas maybe widely spread in the HSZ pore during the local MH formation.

Several mathematical models, which aim at different scientific targets, have been built to quantify and predict the dynamic alternation behaviors of MHSs and their mutual influences with the ambient environment from the last million years to the next thousand years (Archer et al., 2012; Chatterjee et al., 2014; Garg et al., 2008; Liu and Flemings, 2007; Ruppel and Kessler, 2017; Torres et al., 2004; Xu, 2004). The mass and energy transfer has been used to characterize the transformation of biogenic methane production in origin zones, the thermogenic methane migration in deeper seafloor layers, and the MH kinetic reaction in the HSZ. Analogously, these findings have also been used in the study of the Shenhu MHS. Guan et al. (2009, 2014) explained how free gas in the local pores of the HSZ had strongly influenced the formation of MH in the Shenhu seeping environment, and pointed out that in this thermodynamic setting, the speed of MH generation from both dissolved methane and free gas was about four times faster than that by only dissolved methane in aqueous solution. He et al. (2011) designed a 2D model to probe the relationships among local gas chimneys, mud diapirisms, and gas hydrate reservoirs. Su et al. (2014) focused on the sedimentation and water flow rate to simulate the accumulation of hydrate. These numerical investigations have greatly improved the understanding of the Shenhu MHS.

However, some uncertain processes and details about the dynamic formation and accumulation mechanism of the Shenhu MHS still need to be clarified. If methane can be present in the gas phase in the HSZ pore it means that the methane supply is sufficient. Generally, under this kind of gas-aqueous two-phase state, hydrate formation will accelerate more than in only the dissolved methane-liquid one-phase condition, according to the laboratory and field observations (Babu et al., 2013; Egorov et al., 2015; Li et al., 2014; Waite and Spangenberg,

2013; Zhang et al., 2011). Different than previous studies and viewpoints, considering the significant widespread seafloor pathways for fluid migration and the abundance of cold seep marks in the northern SCS (Lu et al., 2015; Shang et al., 2013), the gas-bearing fluids may rapidly flow across the bottom of the local HSZ from below and form the abundant hydrate-bearing sediments within a short period of geological time, driven by some crustal activity. During this rapid MH formation process, the HSZ pore will often transform from two phases to the hydrate-liquid-gas three-phase state. Local sedimentary permeability and capillary pressures may play a negative role on fluid flow and MH aggregation (Nimblett and Ruppel, 2003; Thatcher et al., 2013). Moreover, the heat released by the massive MH formation within a short period time may also result in the rise of the HSZ bottom.

In this paper, we will discuss the possibility of a rapid hydrate-bearing sediment formation mechanism and the corresponding evolution process of the sedimentary properties in the Shenhu MHS. A methane flow-hydrate reaction model has been used to describe this possible geological activity. MH generation from upwardly migrating supersaturated pore fluids is treated as a kind of quasi-equilibrium process. Furthermore, factors such as kinetic reaction, temperature, pressure, permeability of stratum, and fluid fluxes, which evolve in this process of the hydrate-bearing sediment formation and accumulation in Shenhu Area are also investigated.

#### 2. Regional geological setting

The northern slope of the SCS is a passive continental margin and breeds very favorable thermodynamic conditions, sedimentary structures, and methane sources for MH generation. The Shenhu hydrate fields are located in the middle of this area and are seated among the Zhu III Depression to the northwest, the Kaiping-Baiyun depressions to the northeast, and the Xisha Basin to the south (Fig. 1) (GMGS1 2007; Wu et al., 2009). This Shenhu basin has been speculated to have formed in the middle Miocene, and its sediment thickness is generally above 3000 m. Its biomorphic setting is very suitable for the storage and conversion of biogenic methane. A large-scale block body transportation system and deep-water channel system have been widely detected here. These geological structures provide feasible upward migration pathways for pore saturated gas-aqueous mixed fluids. The organic matter content in the HSZ here is about 0.2–1.9%, but relatively larger

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