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Comparison of the characteristics for natural gas hydrate recovered from marine and terrestrial areas in China

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ABSTRACT

China has carried out several drilling campaigns for natural gas hydrate both in marine and terrestrial areas and successfully obtained the samples. The marine gas hydrate samples were firstly recovered from Shenhu area in 2007 and then from the Pearl River Mouth basin in 2013 in South China Sea (SCS). The terrestrial gas hydrate samples were recovered from Qilian Mountain permafrost (QMP) in 2009 and 2013, respectively. In this paper, systematic analyses have been carried out on these gas hydrate samples to compare the characteristics of gas hydrates from SCS with those from QMP. The results indicate that the characteristics of occurrence, structure and gas composition are obviously different. Marine gas hydrate from SCS shows different kinds of occurrence and demonstrates a typical structure I (sI), with cage occupancy of more than 99% methane in large cage and 90% in small cage, respectively, corresponding to hydration numbers of approximately 6.0 by thermodynamic calculation. The guest molecules are predominantly methane (>99%) from biogenic origin produced by CO_2 reduction. However, the terrestrial gas hydrates from QMP occur as a thin layer within the cracks of fine-grained sandstones, siltstones and mudstones, showing a possible structure II (sII) hydrate based on its Raman spectra and gas composition. The molecular composition of hydrate-bound gas indicates that CH_4 only accounts for ~60% of the guests while the others are heavier hydrocarbons (e.g. C_2H_6, C_3H_8 and C_4H_{10}).The cage occupancy ratio of methane in small and large cage (θ_S/θ_L) is around 7.5, suggesting that larger molecules preferentially occupy the large cage of the hydrate. The isotopic analysis shows that hydrate-bound gases in QMP are from thermogenic origin.

1. Introduction

Natural gas hydrate, distributed widely in marine sediments beyond a water depth of 300 m and the terrestrial permafrost, has been considered as a potential energy resource and attracts worldwide attentions (Kvenvolden, 1995; Makogon et al., 2007). So far, totally about 133 hydrate origins have been found in the world, with 123 in the seabed and lake sediment and 10 in the terrestrial permafrost regions. Permafrost hydrates are mainly distributed in Arctic permafrost at high latitude countries such as Russia, the United States and Canada (Collett et al., 2011; Lu et al., 2011a; Max, 2003; Moridis et al., 2005). For example, the first gas hydrate resource was found in Messoyakha, the Soviet Union, in 1960s (Makogon et al., 2007). In 1972, gas hydrate samples were obtained by drilling in the Prudhoe Bay at the north slope of Alaska (Collett, 1993). Since gas hydrates were found in 1971 in Mackenzie delta in Canada, several wells (Mallik L-38, 2L-38, 3L-8, 4L-38, 5L-38) have been drilled and tested for producing natural gas from hydrate, making this region the most extensive research area (Dallimore and Collett, 1999, 2005).

The northern South China Sea (SCS) is a transitional continental margin, which remains lots of features of continental geological structure due to the comparatively short expansion time (Ludmann et al., 2001). This area has experienced intensive neotectonic movements, resulting in complex base structures and complicated fault systems (Wang et al., 2006). In this area, the topography is complex and the slope varies greatly. The water depth is between 800 and 2000 m, with the fathom line roughly parallel to the coast line. The deposition rate was 17.9–19.6 cm/ka and 9.6–14.6 cm/ka for late Pleistocene and Holocene, respectively, somewhat higher than that for Pliocene. The geothermal gradient in the studied area is relatively low (45 to 67.7 °C/km) as compared with other regions (~80 °C/km) in SCS (Wu et al., 2009). The occurrence of gas hydrate in SCS was known as

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postulated from the geophysical and geochemical investigations (Ge et al., 2010; Liu et al., 2011). In June 2007, China Geological Survey organized a gas hydrate drilling campaign, GMGS1, in Shenhu area (Fig. 1a), and obtained the natural gas hydrate samples for the first time in China (Zhang et al., 2007). From June to September in 2013, China's second major expedition, GMGS2, took place in the eastern part of the Pearl River Mouth basin at northeast of the Shenhu area (Fig. 1a). It is found that 9 of the 13 investigated sites contain gas hydrate by combining data from logging and core sampling. A variety of morphologies of gas hydrate, such as massive, laminated, nodular, vein and disseminated, were found in samples retrieved from GMGS2 drilling sites (Zhang et al., 2014).

The unique gas hydrate samples have been obtained in low-latitude mountain permafrost zone, the Qilian Mountain permafrost (QMP) region in Tibet Plateau in northwest China since 2008, although the depth of permafrost layer is relatively thinner(Lu et al., 2011b, 2013; Zhu et al., 2010). Permafrost zone in Qilian Mountains is located in the northern margin of Qinghai–Tibet Plateau, with an area of 10×10^4 km^2 (Fig. 1b). The topography of the drilling area changes from higher in the west and south to lower in the east and north. The altitude is between 4026 m and 4128 m with the permafrost thickness generally 60–80 m. Coal-bed methane is abundant in this area that is beneficial to gas hydrate formation. Some researches indicate that a large number of abnormal signs have been found in this area (Chen et al., 2005; Lu et al., 2007b, 2009). From 2008 to 2014, China Geology Survey organized the "Gas Hydrate Scientific Drilling Project" in QMP in Muri Coalfield. Gas hydrate samples were obtained and well preserved in 2009, and were analyzed in our laboratory for the first time. In August 2013, hydrate samples were obtained again from the DK9, DK11 and DK12 wells, about 450 m in the southeast of the DK2 well. Totally 14 scientific drilling test wells have been carried out and 7 of which successfully obtained the hydrate samples (Fig. 1b).

Analysis of gas hydrate samples can provide directly the information about the characteristics of hydrates and hydrate-bearing reservoirs (Kneafsey et al., 2011). Three crystallographic structures (i.e. structure I (sI), structure II (sII) and structure H (sH)), have been found for natural gas hydrate. The structure depends on the encaged gas component (Davidson et al., 1986; Ripmeester et al., 2005; Uchida et al., 2002). Occasionally, the naturally occurring gas hydrates contain complex gas component (Lu et al., 2007a). For example, gas hydrate recovered from QMP contains only ~60% methane (Zhu et al., 2010), whereas gas hydrates from Shenhu area may contain methane higher than 99% (Liu et al., 2012). The field observation found that gas hydrates from the Pearl River Mouth basin in SCS have various morphologies (Zhang

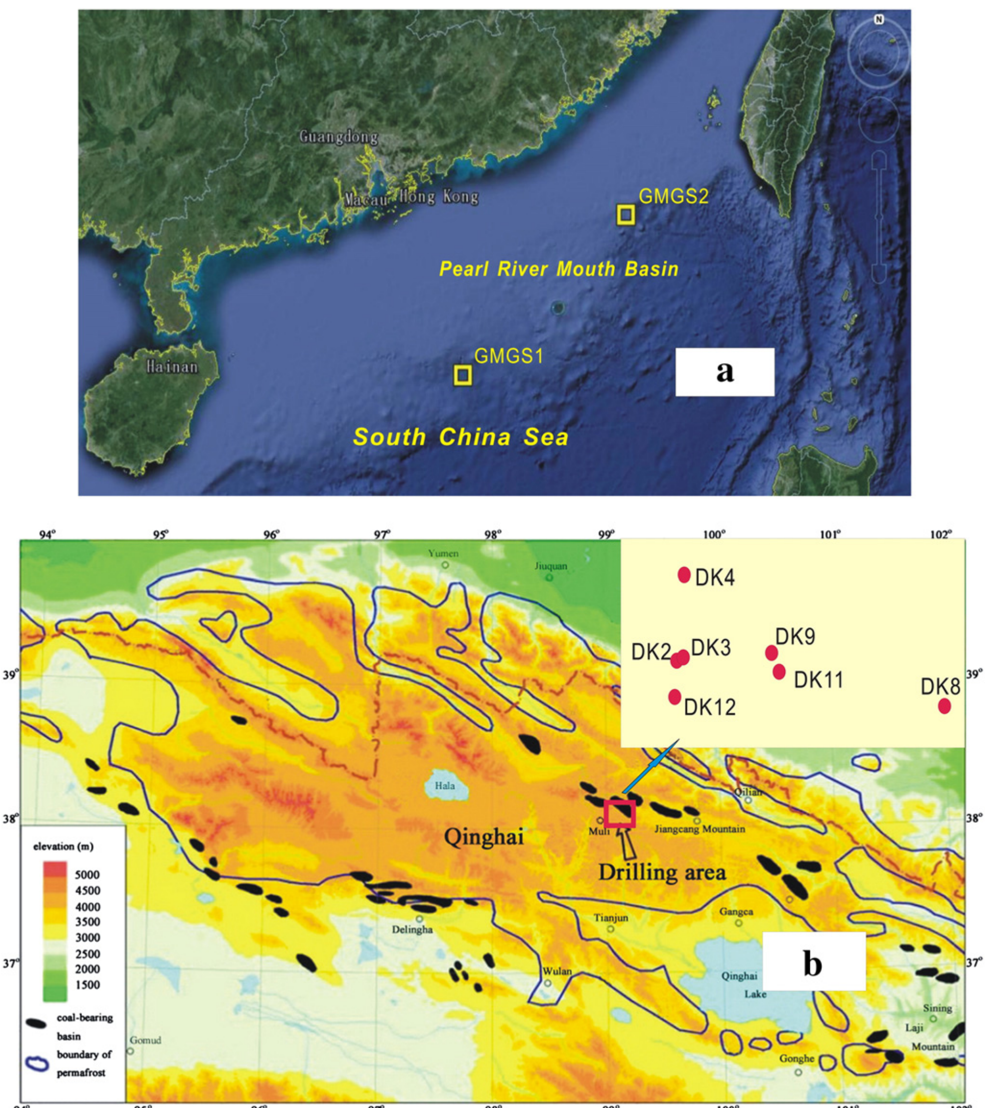

Fig. 1. Map of marine and terrestrial areas of gas hydrate source, showing: (a) locations of GMGS1 and GMGS2 expeditions in SCS and (b) locations of the Scientific Drilling Project of Gas Hydrate in QMP, the enlarged view showing the drilling wells obtained gas hydrate samples (modified from Zhang et al., 2014 and Zhu et al., 2010).