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RESEARCH PAPER

Genetic types of gas hydrates in China

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Abstract: Researches were carried out on the origin of gas hydrate samples from the tundra in the Qilian Mountain, Pearl River Mouth Basin in the northern South Sea and the continental slope of Taixinan Basin in China. Gases of the gas hydrate samples from the Jurassic Jiangcang Formation in the Muli County in Qilian Mountain are mainly of oil-derived origin, characterized by self-generation and self-preservation. $\delta^{13}C_1$ values range from −52.7‰ to −35.8‰, and the $\delta^{13}C_2$ values vary from −42.3‰ to −29.4‰. There was a small amount of coal-derived gases, which might source from the coal-bearing Middle-Jurassic Muli Formation with $\delta^{13}C_1$ of −35.7‰ – −31.3‰ and $\delta^{13}C_2$ of −27.5‰ – −25.7‰. Gases of the gas hydrate samples from the Pearl River Mouth Basin and Taixinan Basin are dominated by bacterial origin of carbonate reduction, with $\delta^{13}C_1$ of −74.3‰ – −56.7‰ and δD_1 of −226‰ – −180‰. A trace amount of thermogenic gases were also found in these basins with $\delta^{13}C_1$ of −54.1‰ – −46.2‰. This study combined the geochemical data of gas hydrates from 20 areas (basins) in the world, and concluded that thermogenic gases of the gas hydrates in the world can be either of coal-derived or oil-derived origin, but dominated by oil-derived origin. A small amount of coal-derived gas was also found in the Qilian Mountain in China and the Vancouver Island in Canada. The coal-derived gas has relatively heavy $\delta^{13}C_1 \geq -45$‰ and $\delta^{13}C_2 > -28$‰, while the oil-derived gas has $\delta^{13}C_1$ from −53‰ – −35‰ and $\delta^{13}C_2 < -28.5$‰. Gas hydrates in the world mainly belong to bacterial origin of carbonate reduction. Methanogensesis of acetate fermentation was only found in some gas hydrates from the Baikal basin in Russia. Bacterial gases of carbonate reduction have relatively heavy $\delta D_1 \geq -226$‰, while gases of acetate fermentation have $\delta D_1 < -294$‰. The bacterial gas of gas hydrates in the world has the highest $\delta^{13}C_1$ value of −56.7‰ and lowest of −95.5‰, with a peak range of −75‰ – −60‰. Gas hydrate in the world has the highest $\delta^{13}C_1$ of −31.3‰ and lowest of −95.5‰ and the highest δD_1 of −115‰ and lowest of −305‰.

Key words: Shenhu area; Qilian mountain permafrost; gas hydrate; biogenic gas; oil-derived gas; coal-derived gas; carbon and hydrogen isotopic composition; genetic type

Introduction

Gas hydrate has been found in the laboratory by chemists for almost 200 years, but its significance in energy has been appreciated for quite a long time. In the 1930s, the oil industry began to pay attention to gas hydrate when it was considered as the cause of the pipeline plugging[1]. In the 1960s, Russian scientists first discovered gas hydrate in the lithosphere[2–3]. Messoyakha gas field, the world's first gas field of gas hydrate, was discovered in the northern west Siberian basin in 1968[4–5]. In the early 1970s, some scientists[6–7] speculated that gas hydrate existed in permafrost and marine sediments, and in the early 1980s, scientists found gas hydrate in sediments surrounding epicontinental sea through offshore deep-water core drilling[8–9]. The Tarm and Eileen gas hydrate reservoirs found in Alaska's north slope region of the United States[10] and the Mallik hydrate accumulation found in Mackenzie Delta tundra[11] confirmed the scientific speculation of the early days made by Stoll et al.[6]. The gas resources in the global gas hydrates are enormous, the estimated reserves of natural resources range from 2.8×10^{15} to 8.0×10^{18} m^3[12], spanning three orders of magnitude. The widely cited global gas hydrate resources are 2×10^{16} m^3 proposed by Kvenvolden[8]. With the increasing energy consumption and pollution in the world, the exploration and development of gas hydrate resources are speeding up. Pilot production of gas hydrate has been carried out in northern Alaska, the Mackenzie Delta, Japan Nankai trough[12] and Chinese Shenhu area in the South China Sea[13].

With a late beginning, China's gas hydrate research and investigation lagged about 30 years behind those in other countries. In the 1980s and 1990s, the relevant units of Ministry of Geology and Mineral Resources, the Chinese Academy of Sciences and the Ministry of Education translated and collected foreign survey and research results on gas hydrates, preparing for the survey of gas hydrate in China. In 1999-2001, Guangzhou Marine Geological Survey (GMGS) took the lead in carrying out high-resolution multi-channel

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seismic survey in the Xisha trough area of the northern South China Sea. In 2002, the national project, the survey and evaluation of gas hydrate resources in China sea areas was officially launched[14]. Thereafter, the research and investigation of gas hydrate were carried out not only in the northern slope of the South China Sea but also in the permafrost regions. In 2008, the gas hydrates drilling in the tundra of the Qilian Mountains made major progress. In terms of pilot production of gas hydrate, over 30.0×10^4 m^3 of natural gas were produced in Shenhu sea area in 60 days from May 10th, 2017 to July 9th, 2017, setting a new world record in gas production time and total volume[13], which outperforms the 20×10^4 m^3 of natural gas produced in 24 days (from June 5th to June 28th) in the Nankai trough.

1. Formation and distribution of gas hydrate

1.1. Formation

Formation of gas hydrate needs four conditions: (1) Low temperature, the optimum temperature is 0 to 10 °C. (2) High pressure, the pressure should be greater than 1.01×10^7 Pa. When the temperature is 0 °C, the pressure should be no less than 3 MPa, equivalent to 300 m hydrostatic pressure. The hydrate can also be formed at higher temperature, usually at a water depth of 300–2 000 m (pressure of 3 to 20 MPa), the hydrate can still form and remain stable at a temperature of 15–25 °C. The upper limit of reservoir formation is the seafloor, the lower limit is about 650 m below the seafloor, and even deeper to 1 000 m below seafloor[14]. (3) Adequate gas source, the sea area with strong contour currents is generally a favorable area for hydrate enrichment because gas source is sufficient in contour currents, for example, the Black Sea hydrate may be related to contour currents effect[15]. The research of gas hydrates in the Northern Slope of Alaska[16] and Canada[17] shows that thermogenic hydrocarbon sources are important for the formation of high abundance gas hydrate. Therefore, gas source is the core factor affecting gas hydrate accumulation and enrichment. (4) Certain amount of water. Water of cage structure is where the gas hydrate stores. Gas and water jointly constitute gas hydrate. Thus, water is one of the important substances in the formation of gas hydrate.

1.2. Distribution

Although the gas hydrate resources are huge, it is controlled by 4 formation factors, and is uneven in distribution. 98% of the global gas hydrate resources that have been found are distributed in the continental slope, only 2% are distributed in the continental polar regions, tundra, inland seas and lakes[18]. China has found geological, geophysical and geochemical evidence of gas hydrate in the deep-water areas of the Xisha Trough Basin, Qiongdongnan Basin, the Pearl River Mouth Basin and the Southwest Taiwan Basin in the northern South China Sea[19], and also found gas hydrates in East China Sea, Eastern Taiwan sea area, Nansha trough and Nansha sea area[18, 20]. Since 2008, several wells have drilled gas hydrate[21–23] from the tundra of Qilian Mountains in Muli region of Qinghai Province. The exploration of gas hydrate in the permafrost region of the Qiangtang basin and the northeastern Mohe region is also positive[20, 24–25].

2. Geochemical characteristics and source of gas hydrate

Gas source correlation and identification is an important supportive study of gas accumulation, migration analysis and resource assessment. Compared with conventional natural gas and even tight gas in unconventional natural gas, 98% of gas hydrate resources are distributed in the ocean, and most of them are biogenic dry gas. Moreover, gas hydrate samples are often lack of scientific information such as heavy hydrocarbon gas and light hydrocarbon gas, adding difficulty to gas source correlation and identification. Research on the gas source can only rely on the low-carbon molecular gas components and their carbon and hydrogen isotope-related parameters. China has obtained samples of gas hydrate in the tundra of Qilian Mountains, the Pearl River Mouth Basin and some blocks of continental slope belt in Southwest Taiwan Basin (Fig. 1), and reported their gas composition, carbon isotope composition. This paper will examine the geochemical characteristics and gas source of these gas hydrates comprehensively.

2.1. Qilian Mountains tundra

The permafrost region of Qilian Mountains is about 10×10^4 km^2, with an annual average temperature of lower than –2 °C, and the thickness of permafrost of 50–139 m[26], which are favorable for gas hydrate formation, so this area has promising exploration prospects[27]. Since 2000, the China Geological Survey has drilled more than 10 gas hydrate scientific wells in Muli Depression of the Southern Qilian Basin, i.e., the Juhugeng mine of Muli coalfield in Muli town, Tianjun County, Qinghai Province located in the southern edge of Qilian Mountains. 11 exploration wells found gas hydrate, namely DK-1, DK-2, DK-3, DK-7, DK-8, DK-9, DK-12, DK13-11, DK12-13, DK11-14 and DK8-19 (Fig. 1). Gas hydrate is mainly stored in siltstone and mudstone of the Middle Jurassic Jiangcang Formation, and secondly sandstone unstable and discontinuous in distribution. The occurrence of gas hydrates is closely related to faults and the burial depth is 133.0–396.0 m[20–23]. Many researchers[20–23, 28–30] have studied the main geochemical parameters of the natural gas from the above wells (Table 1).

2.2. Northern slope of the South China Sea

Guangzhou Marine Geological Survey implemented gas hydrate drilling in 2007, 2013, 2015 and 2016 respectively in the northern slope of South China Sea and found gas hydrate successfully, confirming that the study area is rich in gas hydrate resource. The drillings and investigations have proved that the western part, central part and eastern part of the