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Tectono-thermal evolution and gas source potential for natural gas hydrates in the Qilian Mountain permafrost, China

Yinhui Zuo [a, b, *], Quanfeng Wang [c], Zhenquan Lu [d, **], Hui Chen [c]

[a] State Key Laboratory of Oil and Gas Geology and Exploitation, Chengdu University of Technology, Chengdu 610059, China
[b] College of Energy Resources, Chengdu University of Technology, Chengdu 610059, China
[c] Key Lab of Geomathematics of Sichuan Province, Chengdu University of Technology, Chengdu 610059, China
[d] Oil & Gas Survey, CGS, Beijing 100029, China

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ABSTRACT

The gas source of natural gas hydrates in the Qilian Mountain permafrost was determined based on analyses of the source rocks using model simulations and observed data. The thermal history, maturity history, and hydrocarbon generation/expulsion history of the basin were modelled in an artificial well using the BasinMod 1D software. The modelled geothermal gradient presents a gradual increase from the Permian to the mid-Jurassic (29–51 °C/km) and then a decrease to present day (51–31 °C/km). The mid-Jurassic geothermal fields were shown to play an essential role in the evolution of source rock maturity, with the peak of maturity occurring in the late period of the mid–Jurassic. The Upper Triassic Galedesi Formation is the most important source rock in the South Qilian Basin and experienced two periods of oil generation during the Late Triassic and mid-Jurassic and one period of gas generation during the mid –Jurassic, but only one period of oil and gas expulsion during the mid-Jurassic. The natural gas hydrates in the Qilian Mountain permafrost include relatively high concentrations of methane, ethane and high carbon-number alkanes (e.g., propane, butane); a small amount of heavy hydrocarbons that are heavier than C_6; and some CO_2 and N_2. These compounds are consistent with the characteristics of pyrolysis and wet gas. The gas source of natural gas hydrates in this area was found to come from the deep source rocks of the Upper Triassic Galedesi Formation based on measurements and modelling. This study also reveals that the South Qilian Basin not only contains natural gas hydrates, but may also be a potential oil and gas basin, and hence, further studies may be worthwhile to identify oil and gas resources in the South Qilian Basin.

1. Introduction

Natural gas hydrates (or gas hydrates) are ice-like, clathrate crystalline compounds that are generated from water and gas under high pressure and low temperature in deep-sea sediments or terrestrial permafrost. They have been receiving increased attention as a major new energy resource and an ideal replacement of fossil fuels. Global estimates of their deposits are up to 2.1×10^{15} m^3, which is approximately twice the amount of available conventional coal, oil and gas combined (Makogon, 1982; Kvenvolden, 1988; Makogon et al., 2007). In the 1960s, the former Soviet Union first discovered a gas hydrate reservoir in the Siberian permafrost and developed it in 1969. The total gas production of this reservoir was 5.017×10^9 m^3 within 14 years. The United States began gas hydrate investigations in 1969, and Japan began gas hydrate surveys in 1992. By 2010, surveys and investigations of gas hydrates had been performed in more than 40 countries (Gu, 2010). Recent studies have focused on the formation conditions of natural gas hydrates, as well as experiment simulations and formation process simulation techniques (Kneafsey and Moridis, 2014; Shin et al., 2014; Stern and Lorenson, 2015; Aifaa et al., 2015; Lee et al., 2015; Maeda, 2015; Babakhani and Alamdari, 2015; Govindaraj et al., 2015; Bhade and Phirani, 2015; Baig et al., 2016; Abbasi and Hashim, 2016; Kim and Lee, 2016; Misyura and Donskoy, 2016).

China launched gas hydrate resource surveys and exploration in marine and terrestrial permafrost in 1999. In 2007, gas hydrate

* Corresponding author. State Key Laboratory of Oil and Gas Geology and Exploitation, Chengdu University of Technology, Chengdu 610059, China.
** Corresponding author. Oil & Gas Survey, CGS, Beijing 100029, China.
E-mail addresses: zuoyinhui@tom.com (Y. Zuo), luzhq@vip.sina.com (Z. Lu).

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samples were collected in the Shenhu Area of the South China Sea. In November 2008, gas hydrates were discovered in the Qilian Mountain permafrost (Muli town, Qinghai province, China; Elevation 4062 m). This was the first time anywhere in the world in which a permafrost area of a mid-low latitude plateau was drilled for gas hydrates (Trung, 2012). Gas hydrates were found in the fissures of siltstone, mudstone and oil shale, as well as the pores of sandstone of the Jurassic Jiangcang Formation (Zhu et al., 2010). The discovery of gas hydrates in the Qilian Mountain permafrost motivated studies on sedimentary reservoirs (Wang et al., 2011; Zhu et al., 2010), gas sources (Cao et al., 2009; Wang et al., 2009; Lu et al., 2010,2013; Huang et al., 2011; Liu et al., 2012; Xue et al., 2013), logging responses (Guo and Zhu, 2011), microorganisms (Wu et al., 2012), and gas hydrate types in this region (Meng et al., 2011). However, the gas source of the gas hydrates in the Qilian Mountain permafrost remains unclear. There are currently two major perspectives in terms of the candidates for the gas source: 1) shallow Jurassic coal-bed methane, which is consistent with the temperature and pressure conditions of the gas hydrates (Cao et al., 2009; Wang et al., 2009); 2) deep pyrolytic hydrocarbon, based on the geochemical composition of the gas hydrates (Lu et al., 2010,2013; Huang et al., 2011; Liu et al., 2012; Xue et al., 2013). Nevertheless, no studies have been performed to clarify the gas source of the gas hydrates in the Qilian Mountain permafrost using detailed observations combined with modelling of the physical processes, such as tectono-thermal evolution.

In this study, we aim to assess the gas source of the gas hydrates in the Qilian Mountain permafrost by modelling the thermal history and hydrocarbon generation/expulsion history of different source rocks that are essential for gas generation since the composites of a sedimentary basin are uniquely correlated with the thermal history. Model simulations were validated with observed data to reconstruct the thermal history and hydrocarbon generation/expulsion history in this basin, and multiple potential source rocks were compared and discussed.

2. Geology settings

The South Qilian Basin, which has a total area of 6.3×10^4 km^2, is surrounded by the Qinghai Lake to the east, Hala Lake to the west, Zongwulong and South Qinghai Hills to the south, and mid-North Qilian Mountain to the north, and it has an overall northwest-southeast orientation. It is a residual basin that experienced several stages of post-tectonic movement deformations, including six uplifts and five depressions: the Tianpeng, Datongtian, Yangkang, Baixinghada, Shulinanshan, and Tuolainanshan uplifts and the Tianjun, Xiariha, Muli, Halahu, and Shule depressions (Fig. 1). The strata in this area include a number of formations, such as the Carboniferous formation, Permian Lemengou Formation (P_{1m}), Caodigou Formation (P_{1c}), Hajier Formation (P_{2h}), Triassic Xiahuancang Formation (T_{lx}), Jianghe Formation (T_{1j}), Dajialian Formation (T_{2d}), Qieerma Formation (T_{2q}), Atasi Formation (T_{3a}), Galedesi Formation (T_{3g}), mid-Jurassic Muli Formation (J_{2m}), Jiangcang Formation (J_{2j}) and locally developed Cretaceous and Cenozoic formations. The Qilian Mountain experienced complex tectonic movements throughout history: (1) in the Early Paleozoic, the Qilian Mountain was a small ocean basin between the Qaidam and North China blocks; (2) in the Late Silurian, this ocean basin became closed and underwent uplift and erosion due to the Caledonian movement; (3) in the Carboniferous, this region started to subside and experience a broad shallow shelf deposition; (4) in the Permian, the North Qilian Mountain became land resulting from uplift and the South Qilian Mountain remained as a shallow shelf or epicontinental sea environment due to north-south differential rise and fall; (5) in the Triassic, as the Paleo-Tethys completely closed, the South Qilian Basin deposited sandstone, mudstone and limestone and the entire Qilian Mountain uplifted to land, forming into a denuded area at the end of the Late Triassic because of the Indo-China movement; (6) in the Jurassic, the Early Yanshanian movement caused localized stretching in the Qilian Mountains, creating strips of intermontane faulted basins and depositing coal-bearing clastic rocks in the lacustrine and fluvial environments; (7) in the Cretaceous, Paleogene and Neogene, the Qilian Mountain was dominated by fine red clastic rock and clay stone; and (8) in the Quaternary, the basin was dominated by an ice water-pluvial phase and glacial deposits (Fu and Zhou, 2000).

Multiple sets of potential source rocks were assessed in the South Qilian Basin in Yang (2012) and Wang (2012). In the Jurassic dark mudstone from the Qilian Mountain, the total organic carbon (TOC) reached up to 1.0% and the organic matter was mainly type II kerogen, a compound that is usually derived from planktonic and bacterial remains deposited in marine or continental environments. Type II kerogen is not only a common source of crude oil, but can also yield natural gas with medium to high maturity (0.73%~1.10%). These statistics indicate that Jurassic dark mudstone can be a good source rock in the South Qilian Basin. However, the thickness of the Jurassic dark mudstone layer is less than 100 m (Cao et al., 2009); hence, the hydrocarbon generation potential of the Jurassic dark mudstone is relatively small. In the Lower Permian Caodigou Formation, the TOC is 0.23% and the potential yield ($S_1 + S_2$) is 0.041 mg/g, where S_1 is the amount of free hydrocarbons (gas and oil) in the sample (in milligrams of hydrocarbon per gram of rock) and S_2 is the amount of hydrocarbons generated through thermal cracking of non-volatile organic matter, which is mainly type II kerogen. These values suggest that the Caodigou Formation has little hydrocarbon generation potential. In the Middle Triassic Dajialian Formation carbonate rock, the TOC is 0.05% and the $S_1 + S_2$ value is 0.084 mg/g, with type II kerogen as the main organic matter. Thus, the Dajialian Formation is non-poor source rock, although its hydrocarbon generation potential is small. The Carboniferous dark mudstone is only located in the Muli depression and hence is unlikely to be the source rock. The Upper Triassic Galedesi Formation is located in four depressions, except the Tianjun depression, with TOC of 1.57%, $S_1 + S_2$ of 0.517 mg/g, type II kerogen as the main organic matter and maturity up to 1.37%. Therefore, the Galedesi Formation is the best source rock for hydrocarbon generation of gas hydrates in the South Qilian Basin (Table 1) and was used in this study as the model input.

3. Methods and general parameters

3.1. Methods of thermal history reconstruction

Thermal history reconstruction is of great significance to assess the source rocks and understand the tectonic evolution in a sedimentary basin. Two common thermal indicators, Vitrinite reflectance (R_o) and apatite or zircon fission track (AFT or ZFT) (Waples, 1980; Tissot et al., 1987; Lerche, 1998; Wood, 1998; Sweeney and Burnham, 1990; Crowley, 1991; Hu et al., 2011; Qiu et al., 2004; Zuo et al., 2011,2014,2015a,b,2016; El-Shahat et al., 2009), were used in the analyses of the thermal history reconstruction of the South Qilian Basin.

The paleo-temperature at a certain basin depth is a function of the paleo-heat flow and paleo-burial depth, which are determined by the thermal physical characteristics of the rock (e.g., thermal conductivity, heat production rates). For basins without any hiatus, the paleo-burial depth of the strata can be modelled through back stripping and compaction correction. However, the initial time of tectonic uplift and the extent of erosion of a sedimentary basin are unclear, and the characteristic parameters for paleo-heat flow are