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Hydrocarbon source for oil and gas indication associated with gas hydrate and its significance in the Qilian Mountain permafrost, Qinghai, Northwest China

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ABSTRACT

Since gas hydrate was sampled by drilling in the Qilian Mountain permafrost in 2008, to investigate the source of the gas hydrate and the relationship between gas hydrate and the concomitant oil and gas indication (OGI) have become an important research focus. The rocks bearing gas hydrate and OGI from the Middle Jurassic strata in the Qilian Mountain permafrost, Qinghai, Northwest China were extracted by organic solvent and thermally treated (300 °C and 400 °C) in vacuum glass tubes. The hydrocarbons from the extracts and cracked products of the rocks and the stable carbon isotope of the gas hydrocarbons were studied. The results showed that the OGIs can be classified into two types according to different biomarker characteristics. The I-type of OGIs, which suffered from the process of early biodegradation and later-hydrocarbon input and featured high concentrations of 17 α (H)-diahopane and $\alpha\beta\beta$ -regular steranes, mainly originated from the shallow source rocks of the Middle Jurassic strata, while the II-type one with a series of long-chain alkylnaphthalene may originate from the lower portion of Middle Jurassic or deeper source rocks than the Middle Jurassic strata. The adsorbed gas (300 °C) of the Middle Jurassic rocks was very wet, had a normal carbon isotope sequence, and can be regarded as an organic thermogenic gas derived from Middle Jurassic source rocks. Comparing the adsorbed (300 °C) and cracked (400 °C) gases from the rocks with gases from the gas hydrate and drilling core, we found they had similar stable carbon isotope distributions but different relative contents of methane (C₁), ethane (C₂), and propane (C₃). The Middle Jurassic source rocks are mainly deposited in a freshwater paleo-environment, similar to the parent biomass of the gas hydrate and drilling core gas. The difference of the relative concentrations of C₁-C₃ may result from different formation processes between the adsorbed and cracked gases and the gas hydrate and drilling core gas. The II-type OGI, which possibly originated from deeper strata than the Middle Jurassic, were closely associated with the gas hydrate under the drilling well and had a similar parent biomass and depositional environment as the gas hydrate, showing they have a closely correlated hydrocarbon origin.

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1. Introduction

Gas hydrate is a white crystal formed by water and light gases (such as CH₄, C₂H₆, C₃H₈, i-C₄H₁₀, H₂S, and CO₂) under low

temperature (around 273.15 K) and high pressure (3–5 MPa) conditions (Sloan and Koh, 2008), which is mainly found in deep ocean sediments and terrestrial permafrost zones. Gas hydrate has attracted widespread attention by many researchers since mid-1990 (Moridis et al., 2011; Ruppel, 2011; Lu, 2015) because of its huge energy potential as a new type of clean energy (Milkov, 2004; Boswell and Collett, 2011).

Since gas hydrate was first sampled from wells DK-1, DK-2, and DK-3 in the Qilian Mountain permafrost in 2008 (Lu et al., 2011),

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more gas hydrate samples have been found in wells DK-7, DK-8, DK-9, DK-12, DK13-11, DK12-13, DK11-14, and DK8-19, as well as abnormal phenomenon associated with gas hydrate in some other wells (Lu et al., 2015a), indicating the Qilian Mountains permafrost area has favorable geological conditions, such as sufficient hydrocarbon gas sources, qualified temperature and pressure background, for the formation of gas hydrate (Zhu et al., 2006). The gas hydrate inside which the natural gas has CH₄ content of 54%–76%, C₂H₆ of 8%–15%, and C₃H₈ of 4%–21%, generally occurs in mudstones, oil shales, and sandstones 133–396 m deep (Zhu et al., 2010). Prior studies on gas compositions and their stable carbon isotopes from gas hydrate, drilling core gas, and headspace gas suggested that the gases from gas hydrate were mainly organic thermogenic (Huang et al., 2011; Lu et al., 2013a, 2015a,b; Tang et al., 2015), partially mixed with microbial and thermogenic (Wang et al., 2015), and originated from sapropelic source rock deposited in a freshwater environment (Liu et al., 2012; He et al., 2015). Additionally, inside the rocks from the drilling core bearing gas hydrate in the Qilian Mountain permafrost, Lu et al. (2013a) found a significant amount of oil and gas indication (OGI) phenomena, such as oil trace, oil patch, oil immersion, and oil staining, and thought their presence was closely associated with gas hydrate. These OGIs were mainly composed of intermediate-weighted oil with a small amount of heavy oil, over-heavy oil, and even asphalt, which has been suggested as a sign for gas hydrate occurrence (Lu et al., 2013a,b).

The generation mechanism for the hydrocarbon gases of gas hydrate from the Qilian Mountain permafrost was mainly organic and thermogenic, for which the majority of studies agree. However, the specific origination of the hydrocarbon gases is still unclear because we lack an accurate correlation from gas to source. In essence, the OGIs were closely associated with the gas hydrate and are considered a sign of gas hydrate occurrence, but a study about its hydrocarbon composition and origination has been not reported so far. Therefore, this paper focuses on the geochemical characteristic of OGIs and adsorbed and cracked gases from the rock and carries out the oil (gas)-source correlation based on the hydrocarbon composition and its stable carbon isotope distribution, which is helpful in understanding the relationship between the hydrocarbons from OGIs and gas hydrate and the Middle Jurassic source rocks and the relationship between OGI and gas hydrate.

2. Geological setting

Qilian Mountain is located in the north Qinghai Tibet Plateau and consists of Zoulangan Mountain (North Qilian structural zone), Tuolai Mountain (Middle Qilian block), and the South Qilian structural zone. In the early Paleozoic, the South Qilian basin was a small ocean basin between the Qaidam Block and the North China Block. The ocean basin closed, began to uplift, and suffered erosion under the effect of the Caledonian Movement in the late Silurian (Fu and Zhou, 1998). During the Carboniferous, the basin began to sink and formed a wide shallow-marine shelf facies depositional environment; due to different tectonic evolution in the Permian, the North Qilian basin was uplifted, while the South Qilian basin remained a shallow-marine shelf or epicontinental sea environment; the South Qilian basin was still an ocean basin in the Triassic and deposited a set of marine sandy mudstone and limestone (Fu and Zhou, 1998). However, the whole Qilian area was uplifted when the Paleo-Tethys Ocean closed under the influence of the Indosinian Movement in the terminal of the Late Triassic (Fu and Zhou, 1998). In the early Cretaceous, the South Qilian Block was rapidly cooling and suffered from erosion with the Qilian Mountain uplifting; since the Miocene, the Qilian Mountain has experienced north-eastward rise and growth, forming basin-mountain tectonic landforms (Qi et al., 2016).

The South Qilian Basin, located in the South Qilian tectonic belt, is thought to be rich in oil and gas and can be subdivided into five secondary depressions: Shule Depression, Muli Depression, Halahu Depression, Xiariha Depression, and Tianjun Depression (Fu and Zhou, 1998) (Fig. 1a). In the Muli Depression, there are four good source rocks, referred to as Carboniferous dark mudstone (limestone), Caodigou Formation dark limestone from the Lower Permian, Galedeshi Formation dark mudstone from the upper Triassic, and Jurassic dark shale. Due to these source rocks being excellent in hydrocarbon generation and mature-over mature thermal evolution stage, the South Qilian Basin is regarded as one of greatest potential depressions for oil and gas exploration (Fu and Zhou, 1998).

The study area is located within the Juhugeng mining district, Muli coalfield in the Northwest Muli Depression, administratively belonging to Muli town, Tianjun county in Qinghai Province. This area mainly exposes the Upper Triassic, Quaternary, and Middle Jurassic strata (Fig. 1b). The Middle Jurassic strata (Fig. 2), a coal-bearing clastic rock, consists of a suite of fluvial, lacustrine, and swamp facies sediments (Wen et al., 2006). The Middle Jurassic source rock from the Juhugeng mining district included the Muli Formation (J_{2m}) and the Jiangchang formation (J_{2j}) from bottom to top. The Muli formation is mainly a braid-shaped river, lacustrine, and swamp facies sediment, with sandstone and coal line, while the Jiangchang Formation is a delta-lacustrine facies sediment and contains mudstone, oil shale, and sandstone (Lu et al., 2015a,b; Tang et al., 2015). The gas hydrate and abnormal phenomenon associated with gas hydrate were found in the Muli Formation and the Jiangchang Formation (Lu et al., 2010, 2013a,b).

3. Materials and experiment

3.1. Samples and their pre-treatment

Rock samples, including sandstone, mudstone, oil shale, and limestone, were collected from the Middle Jurassic strata bearing gas hydrate from wells DK9, DK10-16, DK11-14, DK12-13, and DK13-11 in the Qilian Mountain permafrost area (Figs. 1b and 3). The five wells are all located at the Sanlutian bare field of the Juhugeng Coalmine Area in Qinghai Province. The horizontal distance between these wells is very small, with a maximum distance around 1 km (Lu et al., 2015a). Details about the well sites are in Fig. 1b.

Sandstone samples marked DK9-O-01, DK9-O-09, DK9-O-18, and DK9-O-19 from DK9 well, bear OGI phenomena such as oil trace, oil patch, oil immersion, and oil staining. The other samples from DK9 well were mudstones and oil shales and used for releasing adsorbed and cracked hydrocarbons from the rock. The samples of mudstone, argillaceous sandstone, or sandy mudstone in wells DK10-16, DK11-14, DK12-13, and DK13-11 were used for releasing cracked gas from kerogen or residual organisms. A previous study showed that the source rocks from wells DK2 and DK3 in the Qilian Mountain permafrost basically did not undergo thermal pyrolysis at 300 °C but would release cracked gas at 400 °C when they were warmed in the closed glass tube systems (Lu et al., 2013c). Therefore, the temperatures 300 °C and 400 °C are selected for heating the samples in the closed glass tube systems to release the adsorbed and cracked gases.

All rock samples are milled into powder and then treated using the following steps: (1) the mudstone and oil shale samples are analyzed by utilizing a Rock-Eval 6.0 Standard Pyrolysis Analyzer to obtain the basic pyrolysis data. (2) All samples are extracted for 120 h with dichloromethane to obtain the soluble fractions of the rocks. The residues are pyrolyzed at 400 °C for 72 h in the closed glass tube systems, obtaining cracked gas from kerogen or residual organic matter. (3) The soluble components from the DK9 well

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