Chemical Geology 352 (2013) 70–80

Contents lists available at SciVerse ScienceDirect

Chemical Geology

journal homepage: www.elsevier.com/locate/chemgeo

Timing of biogenic gas formation in the eastern Qaidam Basin, NW China

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ARTICLE INFO

Article history:
Received 25 March 2013
Received in revised form 23 May 2013
Accepted 2 June 2013
Available online 11 June 2013

Editor: David R. Hilton

Keywords:
Biogenic gas
Stable isotope
Formation water
Chlorine-36 dating
Qaidam Basin

ABSTRACT

Quaternary biogenic gas from the Qaidam Basin, NW China is regarded as one of the representative examples of early generation biogenic gas (gases formed by microbial activity during sedimentation of source rocks). However, new age determinations of natural gas and formation waters make this generally accepted assumption questionable. Gas in the Qaidam Basin is dominantly methane (>99.0%), and has $\delta^{13}C_{CH_4}$ values in the range of −68‰ to −65‰, reflecting the microbial origin. Carbon and hydrogen isotope values of methane, coupled with carbon isotope values of carbon dioxide and dissolved inorganic carbon (DIC), reflect water derived hydrogen in the methane and a dominant CO_2 reduction metabolic pathway for the formation of biogenic methane. Coproduced formation waters have hydrogen and oxygen isotope values similar to surface water from the Golmud and Ortmuren rivers (*T-test* = 0.75), but different from the original formation water (*T-test* < 0.01) on the basis of the δD values of the long-chain n-alkanes, indicating open hydrologic communication between meteoric water and the reservoir. The analyzed waters that appear to be the source of hydrogen during methanogenesis are at least 1 to 2 Ma younger than the host reservoir sediments, inconsistent with an early gas generation hypothesis. The apparent age of 1.34 Ma to 0.35 Ma BP for formation waters suggests that the earliest accumulated biogenic gas was formed shortly after deposition of the K_1 sequence, around 1.35 Ma BP. This corresponds to a regional uplift, erosion, and water influx event. Economic accumulations of biogenic gas were thus most likely generated "late" in the Qaidam Basin, rather than at the same time as sediment deposition. Fresh water recharge may be critical to enhancing biogenic methane generation by introducing nutrients and/or microbes into the reservoir.

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1. Introduction

Subsurface microbial activity occurs whenever conditions in the rocks permit microorganisms to thrive (Rice and Claypool, 1981). Widespread discoveries of microbial communities in the subsurface have led to speculation that biogenic gas (derived from microbial activity) may comprise a larger component of natural gas reserves than previously thought (Kotelnikova, 2002; Milkov, 2011). The determination of timing of biogenic gas formation is very important in order to probe the deep biosphere, and to understand the processes involved and controlling factors of gas generation. As well, different timing of gas generation would require different exploration and development strategies of commercial biogenic gas fields (Shurr and Ridgley, 2002).

Biogenic methane is a common feature in recent sediments, and can make a significant contribution to gas production in highly prolific gas basins (Rice and Claypool, 1981; Schoell, 1983; Rice, 1993; Scott et al., 1994; Martini et al., 1998; McIntosh et al., 2002; Head et al., 2003; Faiz and Hendry, 2006; Strąpoć et al., 2007; Flores et al., 2008; Warwick et al., 2008; Grasby et al., 2009; Zhang et al., 2011; Shuai et al., 2013). However, gas generation (methanogenesis) and charge time have rarely been addressed in biogenic gas systems. Unless specific information is available about the stratigraphic location, age and burial history of potential gas source beds, meaningful predictions about the timing of hydrocarbon fill relative to reservoir and trap development may be very difficult. The timing of biogenic gas generation is generally controversial in known systems. Shurr and Ridgley (2002) proposed that there can be two end members of early or late biogenic gas generation. The former is initiated shortly after deposition of the source and reservoir rocks, while the later occurs long after deposition. Rice

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0009-2541/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.chemgeo.2013.06.001

(1993) noted that most biogenic gas was generated early in the burial history. In contrast, Clayton (1992) suggested that because seals are inadequate in unconsolidated muds of basins with extremely high deposition rates, most early generated shallow biogenic methane escaped before deep burial and seal development. However, no reliable dating data have been provided in either of these case histories. On the bases of isotopic composition of authigenic carbonate, fluid inclusion, and burial history data, Fishman et al. (2001) inferred that biogenic gas in southwestern Saskatchewan and southeastern Alberta can be regarded as an early gas generation system from the time of sediment deposition (about 85 Ma) until about 65 Ma. A similar conclusion was drawn by Lillis (2007), on the basis of a hydrological study and isotopic analysis of formation water. On the other hand, Noble and Henk (1998) suggested that large accumulations of biogenic methane are not necessarily restricted to early entrapment from freshly deposited marine sediments, on the basis of the Terang–Sirasun case study. Their study shows that traps may be filled with biogenic gas long after initial deposition of source beds, provided that the required conditions for active methanogenesis are maintained throughout this period. Biogenic gases from coals and organic rich shales have also been regarded as late-generation system in several studies (Scott et al., 1994; Martini et al., 1998; McIntosh et al., 2002, 2010; McIntosh and Walter, 2005; Formolo et al., 2008; Klein et al., 2008; Grasby et al., 2009, 2012; Bates et al., 2011; Schlegel et al., 2011a, 2011b). Biogenic gas has also been found to have a late generation origin due to methanogenic biodegradation of petroleum, including 1700 tcf of dry gas in the West Siberian Basin (Milkov, 2011).

The Quaternary strata in the Qaidam Basin, China, contain the youngest commercial biogenic gas accumulation in the world, with a proven reserve of 320×10^9 m^3 (Pang et al., 2005; Dang et al., 2008). It has been regarded as a typical early generation system due to the obvious short depositional time and young age of the strata (e.g., Gu, 1993; Zhou et al., 1994; Pang et al., 2005; Dang et al., 2008). However, we show here, on the bases of gas and coproduced water chemical and isotopic compositions, hydrogeologic setting, precise biostratigraphic age and burial history analysis, that the early generation assumption is questionable.

2. Geology of study area

The Qaidam Basin, located at the northern edge of the Qinghai–Tibet Plateau, is a large Mesozoic–Cenozoic continental basin, bounded by three major mountain ranges (i.e. Kunlun, Qilian and Altun mountains, Fig. 1a). Based on the basement structure and sedimentary sequence, this basin can be divided into three first order structural units: the northern margin fault-fold belt, and the western, and eastern depressions.

Quaternary fluvial–lacustrine sediments were mainly deposited in Sanhu area of the eastern depression due to uplift in the west and consequent eastward shift of the depocenter (Fig. 1b). Quaternary sediments, that comprise the Qigequan formation mostly range from 1500 to 2500 m thick (greatest thickness = 3400 m), and have an average depositional rate around 700 to 800 m/Ma. The Qigequan formation can be divided into 12 units (K_0–K_{13}, from top to base) on the bases of seismic reflectance features (Fig. 1c). Lake development reached its climax by the middle Pleistocene (units K_6–K_7), with the deposition of dominantly deepwater fine-grained argillaceous sediments. Several thin layers of halite, with variant thickness from 0.1 m to 2 m, occurred from K_{13} to K_0. The thickest salt layers occur in K_{10}–K_9, K_6–K_7 and K_0. In the late Pleistocene, the Sanhu area was gradually elevated due to the influence of the Quaternary neo-tectonic movement. About 500 m has been eroded along the Sebei–Tainan gas fields during the Holocene, while slightly less sediment has been eroded in the southern part of the depression (Gu, 1993) (Fig. 1d).

The sediments consist mainly of mudstone and sandy mudstone interbedded with siltstone, argillaceous siltstone and carbonaceous mudstone. The organic input is predominately from terrigenous herbaceous and low shrubbery plants. Two types of source rock were developed in the Sanhu area. The organic rich one is carbonaceous mudstone with TOC up to 30% and thickness from 150 to 200 m (Zhou et al., 1994), while the organic poor one is argillaceous mudstones with an average TOC of 0.3% and over 2000 m thick (Gu, 1993). The measured vitrinite reflectance values for Quaternary sediments buried less than 2000 m is below 0.5%Ro, implying an immature nature (Dang et al., 2008).

The Quaternary gas reservoirs in the Sanhu area were deposited mainly near shoreline, in shallow lake settings, with the reservoir rocks being primarily coastal silts and sheet silts. The lithology is dominated by silty detritus, with siltstones and argillaceous siltstones accounting for over 90% of the reservoir volumes. Stacked reservoirs have large cumulative thickness (200 to 300 m), high vertical heterogeneities, and good horizontal connectivity. Because all of the Quaternary reservoirs are currently at the early diagenesis stage, they possess excellent primary porosity and permeability (Dang et al., 2008).

Up to now, six biogenic gas fields have been discovered, with proven reserves of 320 billion cubic meters. Sebei 1, 2 and Tainan gas field are three largest fields and each field has nearly one hundred billion cubic meters of proven reserve. The gas plays discovered in the Sanhu area are mainly within the Quaternary which serves as both source and reservoir rock. The gas reservoir can be defined as a continuous-type accumulation since no unified gas–water contact can be recognized. Gas accumulation is not only controlled by the scale and locality of the gas source rock, but is also closely related to the seal integrity, the location of structural/stratigraphic traps, and the hydrogeological regime of the groundwater systems.

3. Sampling and experimental methods

Eleven natural gas and coproduced formation water samples were collected from producing gas wells completed in the Sebei 1 and Sebei 2 gas fields. Three additional water samples were collected from the Dabsan Lake, Golmud River and Ortmuren River. The gases were stored in a steel cylinder. All water samples were collected directly from the well head in a Nalgene carboy. After pH and temperature were measured, samples were filtered with a 0.45 μm filter and preserved immediately in the field. All sample containers were filled with no headspace, kept on ice in the field, and stored at 4 °C in the laboratory until analyzed.

Gas composition analysis was performed on an HP6890 gas chromatograph (GC), with a precision of ±1%. Stable carbon and hydrogen isotopic analysis of natural gas was performed on a gas chromatograph combustion isotope ratio mass spectrometer (GC-C-IRMS) with a Thermo-MAT 253 mass spectrometer, and a Thermo Delta V Advantage mass spectrometer, coupled to a Varian 6890 capillary GC at the State Key Laboratory of Geochemistry CNPC, with precisions of 0.2‰ and 3‰, respectively.

The alkalinity of water samples was determined by titration within 24 h after sampling via the Gran-Alk method (Gieskes and Rogers, 1973). Major cations (Na, K, Ca, Mg) were analyzed (precision 5%) with an atomic absorption spectrometer (AA-6800), and major anions (Cl, SO_4) were analyzed (precision 5%) following standard methods (China SY/T 5523, 2006). Br was determined via ion chromatography. Stable oxygen and hydrogen isotopic ratios of formation water ($^{18}O/^{16}O$, D/H) were measured on a Finnigan Thermocouple Elemental Analyzer (TC/EA) on-line with a continuous flow Finnigan MAT 253 IRMS at the Laboratory of Resources and Environments, Xi'an University of Technology, with the precisions of ±0.1‰ and ±0.5‰ (SMOW = standard mean ocean water), respectively. Stable carbon isotope ratios of total dissolved inorganic carbon were measured on CO_2 gas extracted with H_3PO_4 after introducing 10 mL of water under a vacuum into the measurement line on a Finnigan-MAT 251 Delta mass spectrometer at the State Key Laboratory of Loess and

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