Marine and Petroleum Geology 91 (2018) 532–549

Contents lists available at ScienceDirect

Marine and Petroleum Geology

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

Research paper

Origin of deep sour natural gas in the Ordovician carbonate reservoir of the Tazhong Uplift, Tarim Basin, northwest China: Insights from gas geochemistry and formation water

Yangyang Wang[a,b], Jianfa Chen[a,b,*], Xiongqi Pang[a,b], Baoshou Zhang[c], Zheya Chen[a,b], Guoqiang Zhang[a,b], Guangping Luo[a,b], Liwen He[a,b]

[a] State Key Laboratory of Petroleum Resources and Prospecting, China University of Petroleum, Changping District, Beijing, 102249, China
[b] College of Geosciences, China University of Petroleum, Beijing, 102249, China
[c] Research Institute of Exploration and Development, Tarim Oilfield Company, PetroChina, Korla, Xinjiang, 841000, China

ARTICLE INFO

Keywords:
Sour natural gas
TSR
Formation water
Carbon isotope ratio
Ordovician
Tarim Basin

ABSTRACT

Deep sour gases are found in the Ordovician carbonate reservoir of the Tazhong Uplift in the Tarim Basin, northwest China. All sour gases in this field are dominated by gas hydrocarbons (C_1–C_4) with varying amounts of H_2S. Most of the gases in the eastern Tazhong are characterized by high gas dryness coefficients (> 0.95), ^{13}C-enriched methane ($\delta^{13}C_1$ ranging from −44.6‰ to −35.8‰), and small difference between values of the $\delta^{13}C_2$ and $\delta^{13}C_1$ ($\delta^{13}C_{2\text{-}1}$). In contrast, the gases in the western Tazhong are characterized by relatively low gas dryness coefficients (mostly < 0.90), ^{13}C-depleted methane ($\delta^{13}C_1$ ranging from −54.9‰ to −35.7‰), and high values of $\delta^{13}C_{2\text{-}1}$, indicating relatively lower maturity. Gas compositions, carbon isotope ratios, and diamantane data together with thermal pyrolysis experiments using sealed gold tubes suggest that the Ordovician gases in the Tazhong Uplift belong to oil cracking gases mixed with high-maturity ^{13}C-enriched dry gases, which originated from the deeper paleo-reservoirs cracking. The H_2S (0.0008%–23.1000%) is attributed to thermochemical sulfate reduction (TSR); in contrast to typical TSR-altered gases, the natural gases in the western Tazhong are characterized by low dryness coefficients, relatively ^{13}C-depleted methane, and high $\delta^{13}C_{2\text{-}1}$, suggesting a low degree of TSR alteration. Observations suggest that the H_2S contents are generally high in the western Tazhong, with positive correlation to Mg^{2+} concentrations and total dissolved solids (TDS) in the Ordovician formation water, indicating that TSR has entered early period of SO_4^{2-} contact ion pairs (CIPs) oxidation. Compared with the eastern Tazhong, the Mg^{2+} concentrations and TDS in the western Tazhong are higher and form more CIPs that accelerate TSR and produce H_2S. In addition, based on the analysis of the Cambrian formation water and the Cambrian gypsiferous salt, TSR might have also exist in the deeper Cambrian reservoirs, and the TSR-altered gases with high content of H_2S migrated along faults to Ordovician reservoirs.

1. Introduction

Currently, petroleum exploration in China has shifted to deep and super-deep gas accumulations. Consequently, the generation, evolution and conservation of the deep gases have become the current hotspots and difficulties in the field of oil and gas geochemistry have spurred investigations (Zhu and Zhang, 2009; Wang et al., 2012). Due to the fact that gas isotope ratio and composition convey important information regarding the gas genesis and post-genetic evolution (Scott et al., 1994; Prinzhofer and Huc, 1995; Milkov, 2005, 2010), in addition to compositional data such as dryness coefficients and nonhydrocarbon components, stable isotope ratios are fundamental parameters for assessing the origin of natural gas and hydrocarbon evolution. According to the previous published studies, the genetic elements (Schoell, 1983, 1988; Chung et al., 1988; Clayton, 1991; Scott et al., 1994; Whiticar, 1994, 1999; Berner et al., 1995; Prinzhofer and Huc, 1995; Rooney et al., 1995; Guo et al., 2009; Pan et al., 2010; Milkov et al., 2007; Milkov, 2011) including gas sources and their degree of thermal evolution are the important factors controlling the compositional and isotopic characteristics of hydrocarbon gases. Consequently, series of methods have been proposed to identify the origin of the gases and the possible processes of gas generation (Dai et al., 1986; Chung et al., 1988; Rooney et al., 1995; Huang et al., 1999). In addition to the genetic processes, the isotopic and molecular signatures are controlled

* Corresponding author. State Key Laboratory of Petroleum Resources and Prospecting, China University of Petroleum, Changping District, Beijing, 102249, China.
E-mail address: jfchen@cup.edu.cn (J. Chen).

https://doi.org/10.1016/j.marpetgeo.2018.01.029
Received 25 August 2017; Received in revised form 18 January 2018; Accepted 22 January 2018
0264-8172/

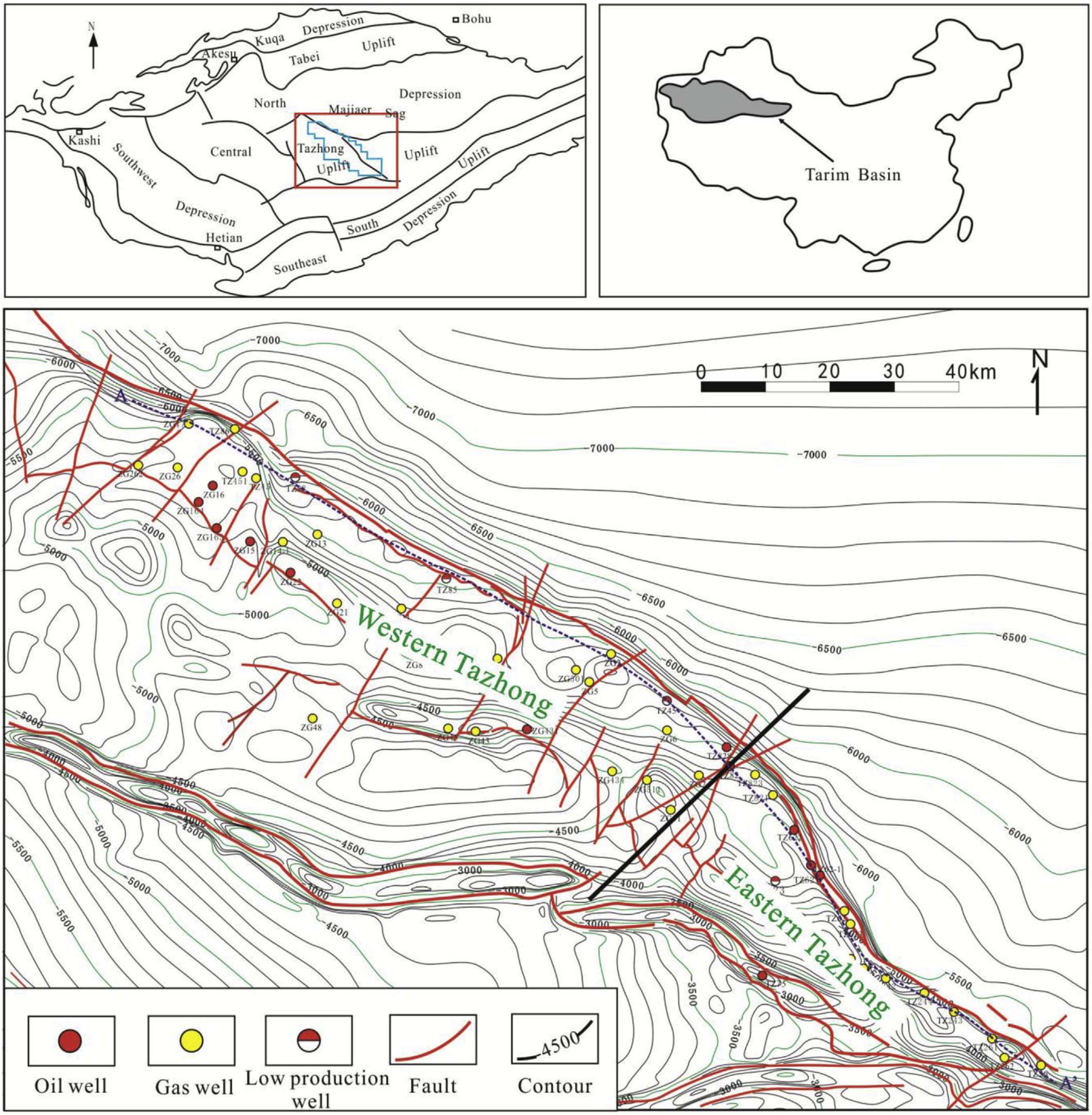

Fig. 1. Ordovician reservoired oil-gas distribution in the Tazhong Uplift, Tarim Basin, NW China.

by the post-genetic processes (Prinzhofer and Huc, 1995, Krooss, 1988; Krooss et al., 1992a, b, Friedrich and Juntgen, 1972; James, 1990; Whiticar, 1994, 1999; Pan et al., 2006; Lu et al., 2010; Dai et al., 2004; Burruss and Laughrey, 2010). It has been proposed that deep hydrocarbons undergo thermochemical sulfate reduction (TSR) that changes the compositional and isotopic characteristics of natural gases (Xiao, 2014). In deep gas reservoirs, sour gas with high H_2S content is produced from complex generation and accumulation processes. TSR is considered to produce H_2S, whereby sulfate minerals and oil react at high temperature over 120 °C (Orr, 1977; Krouse et al., 1988; Sassen, 1988; Worden et al., 1995; Machel et al., 1995; Machel, 2001). Secondary alteration of deep natural gas, including TSR, adds to the complexity of explaining the gas formation and evolution and hinders the reconstruction of the gas evolution process. Thus, it is critical to clarify the origin and secondary alteration of sour gas.

Many studies have described the isotopic and compositional characteristics of TSR-altered gases and TSR in detail (Orr, 1977; Worden et al., 2000; Anderson and Garven, 1987; Heydari and Moore, 1989; Claypool and Mancini, 1989; Heydari, 1997; Manzano et al., 1997; Bildstein and Worden, 2001; Cai et al., 2003, 2004; Zhang et al., 2005, 2007, 2008b; Zhu et al., 2005, 2010a, b). Subsequently, it is suggested that the reservoirs mainly comprise anhydrite that provides the reactants for TSR of heavy hydrocarbons and increases the dryness coefficients ($C_1/\Sigma(C_1–C_5)$, the relative abundance of CH_4 among CH_4, C_2H_6, C_3H_6, n-C_4H_8, i-C_4H_8 and C_5H_{12}). In addition, residual hydrocarbon gas becomes enriched in ^{13}C during TSR owing to the kinetic