



Research paper

TSR-altered oil with high-abundance thiaadamantanes of a deep-buried Cambrian gas condensate reservoir in Tarim Basin



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ABSTRACT

The first exploratory well, the ZS1C well, with 158,545 m³ daily gas production was discovered in 6861–6944 m deep strata of the Cambrian gypsolyte layer of the Tarim Basin, China in 2014. The discovery opens a new target for the Cambrian-reservoired oil and gas exploration, and directly leads to large-scale oil and gas exploration of the deep-reservoired Cambrian oil and gas fields in the Basin. Comprehensive two-dimensional gas chromatography/time-of-flight mass spectrometry and a comprehensive two-dimensional gas chromatography–flame ionization detector revealed the presence of abundant adamantane compounds, 2-thiaadamantanes and 2-thiadiamantanes, and a large amount of sulfur-containing compounds in the condensate oil. The formation of organic sulfur-containing compounds, such as 2-thiaadamantanes, is an indication of sulfur incorporation from the gypsum in the stratum into oil and gas in the course of TSR. This reservoir has apparently suffered severe TSR alteration because (1) High content of H₂S, (2) H₂S sulfur isotopes, (3) CO₂ carbon isotopes, and others abundant data to support this findings. Similar sulfur isotopic composition of H₂S, oil condensate and the gypsum in the Cambrian strata indicate that the produced condensate is experienced TSR alteration. Therefore, the deep-accumulated Cambrian oil reservoir has experienced severe TSR alteration, and accumulated natural gas and condensate contains high sulfur content.

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1. Introduction

As a type of rigid polymeric and cyclic hydrocarbon compound with a structure similar to diamond, the abundance of adamantane is often high in high-maturity crude oil and condensate (Dahl et al., 1999). In the cracking of crude oil at high temperature condition, adamantane-containing compounds may be constantly generated and accumulated (Wei et al., 2007). The presence and abundance of adamantane is often used to measure the cracking degree of crude oil (Dahl et al., 2003; Wei et al., 2007; Zhang et al., 2011). However, thiaadamantane is the compound generated in the process when the carbon atom in the adamantane molecules is replaced by sulfur atom. At present, this compound has been detected in the deep-accumulated crude oil in the Bon Secour Bay in the Mobile Bay gas field, located offshore Alabama in the northern Gulf of Mexico, USA (Wei and Mankiewicz, 2011), the Jurassic Smackover (USA) and the Devonian Nisku (Canada) Formations (Hanin et al., 2002). It is believed that thiaadamantane is the indicator product of the

thermochemical sulfate reduction (TSR) between hydrocarbons and sulfates under high temperature in deep-buried strata (Wei et al., 2012).

TSR is a reservoir alteration process in which petroleum hydrocarbons are oxidized by inorganic sulfate, ultimately yielding CO₂ and H₂S (Orr, 1974; Krouse et al., 1988; Worden et al., 1995; Machel et al., 1995; Wei et al., 2007; Zhang et al., 2008a). TSR is well documented in numerous geological settings from around the world (Claypool and Mancini, 1989; Heydari and Moore, 1989; Rooney, 1995; Worden and Smalley, 1996; Carrigan et al., 1998; Machel, 1998, 2001; Worden et al., 2000; Cai et al., 2003, 2004; Li et al., 2005; Hao et al., 2008; Zhu et al., 2011). It is widely believed that the cause for high hydrogen sulfide content in gas and oil reservoir is from TSR (Machel, 2001; Zhu et al., 2010, 2015a). Hydrogen sulfide is generated by the reaction between hydrocarbons and sulfates driven by thermal chemical sulfate reduction. A large amount of excellent research work has identified the trigger mechanism, reaction condition, and dynamic mechanism of TSR (Zhang et al., 2007, 2008b; 2012; Ma et al., 2008; Ellis et al., 2011; Amrani et al., 2012). TSR involves the participation of various hydrocarbons, which have different reactivities because of their

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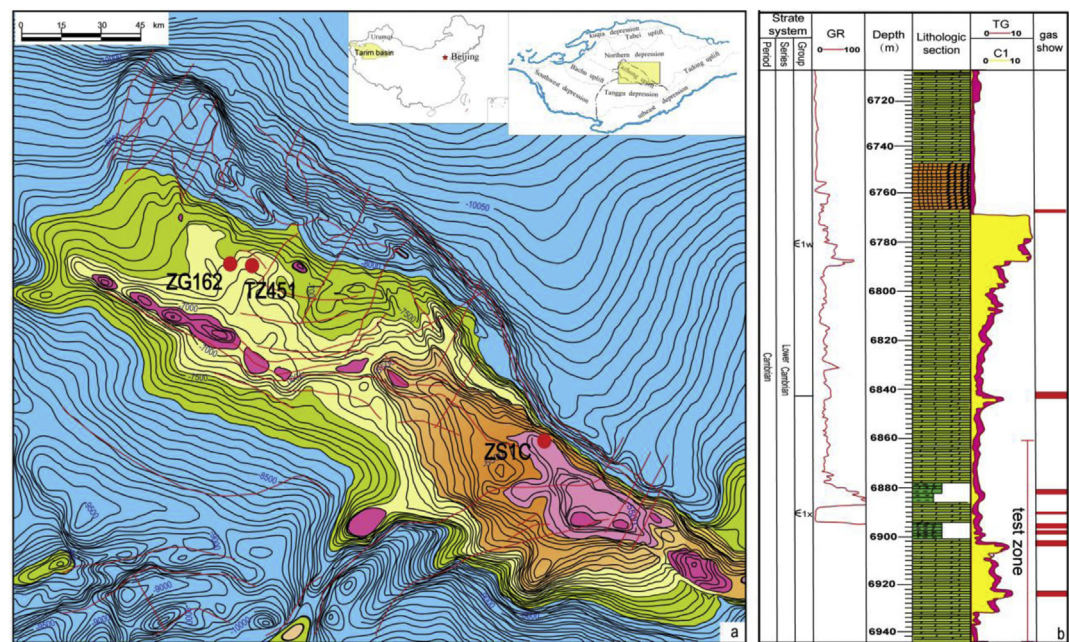


Fig. 1. Structural map of the top Cambrian in the Tazhong area (a) and the lithology column for the Cambrian oil and gas reservoir of ZS1C well (b).

differences in chemical structures and sulfur contents. The quantity and type of the intermediate products of the reaction are hard to estimate due to the participation of different hydrocarbons (Cai et al., 2005; Zhang et al., 2005; Worden and Cai, 2006; Zhang et al., 2006; Zhu et al., 2005, 2015b). TSR reaction can result in complex changes in the fluid components of oil and gas reservoirs by producing some new sulfur-containing compounds (Jiang et al., 2008; Wei et al., 2012). Therefore, oil and gas reservoirs altered by TSR reaction could contain some unique geochemical features, which could indicate the presence/absence of TSR, the intensity or extent of TSR. In recent years, the development of two-dimensional gas chromatography/time-of-flight mass spectrometry (GC × GC–TOFMS) has provided detection means for fast and accurate identification of new compounds. Stable isotopes of carbon and sulfur have proven to be particularly successful diagnostic tools for monitoring the extent of TSR (Amrani et al., 2008, 2012). These analytical means can help identify whether TSR occurs in the deep-buried oil and gas reservoir, its alteration intensity, and the distribution of hydrogen sulfide for risk assessment.

The Tarim Basin is the largest oil-and-gas-bearing basin in China that covers an area of roughly $56 \times 10^4 \text{ km}^2$. It is a typical superimposed basin with lower Paleozoic carbonate rocks containing rich oil and gas resources. Having been buried underground at a great depth for a long period of time and experiencing multi-cycle superimposition and transformation, the oil and gas distribution is highly complex (Zhao et al., 2009; Wang et al., 2013; Zhou, 2013; Zhang et al., 2014; Zhu et al., 2012). The ZS1C well was drilled in the middle of the central uplift of the Tarim Basin (abbreviated as Tazhong area), and the daily gas production is $158,545 \text{ m}^3$ in

6861–6944 m deep strata of the Cambrian gypsolyte layer. This is the first important discovery of commercial oil and gas accumulations in the Cambrian reservoirs in the Tarim Basin (Wang et al., 2014) (Fig. 1), leading directly to the exploration of a new stratum under current breakthrough in the ZS1C. Intensive oil and gas exploration work in the deep-buried Cambrian system in the Tarim Basin has been initiated since then, and the depth for oil and gas exploration has been extended to 7000–10,000 m. In the present study, molecular and sulfur and carbon isotopes were analyzed on gases and condensates using GC × GC–TOFMS and GC × GC–FID to identify whether or not and to what extent of TSR alteration occurred in this deep Cambrian reservoir.

2. Materials and methods

2.1. Instruments

The comprehensive GC × GC system for the GC × GC–TOFMS is from Leco Corporation. Studies reporting GC × GC analysis of condensate samples are rare (Li et al., 2008). The GC × GC system was composed of an Agilent 7890 GC coupled to a hydrogen flame ionization detector (FID) and a liquid-nitrogen-cooled pulse jet modulator. The TOF mass spectrometer is a Pegasus 4D (Leco Corporation). All the data were processed with ChromaTOF software.

2.2. Analytical method for GC × GC–TOFMS

The one-dimensional chromatographic column was a DB-petro (50 m × 0.2 mm × 0.5 μm). The temperature program used was

Table 1
Physical properties of the Cambrian crude oil in Well ZS1C.

Depth (m)	Density (g/cm ³)		Viscosity (mPa s)		Wax content (%)	Sulfur (%)	Resin content (%)	Asphaltene (%)	Solidification point (°C)
	20 °C	50 °C	20 °C	50 °C					
6861–6944	0.9300	0.9108	2.382	2.170	2.80	2.06	0.29	0.18	–30
	0.9298	0.9106	2.345	2.136	4.9	2.67	0.19	0.43	–30
	0.9274	0.9081	2.401	2.181	0.20	2.68	0.45	0.08	–30

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