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Anomalies of natural gas compositions and carbon isotope ratios caused by gas diffusion – A case from the Donghe Sandstone reservoir in the Hadexun Oilfield, Tarim Basin, northwest China

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ABSTRACT

Natural gases in the Carboniferous Donghe Sandstone reservoir within the Block HD4 of the Hadexun Oilfield, Tarim Basin are characterized by abnormally low total hydrocarbon gas contents (< 65%), low methane contents (< 10%) and low dryness coefficients (< 0.5), and a reversal of the normal trend of carbon isotope ratios, showing $\delta^{13}C$ methane (C_1) > $\delta^{13}C$ ethane (C_2) < $\delta^{13}C$ propane (C_3) < $\delta^{13}C$ butane (C_4). Specifically, methane is enriched in ^{13}C with the variations in $\delta^{13}C_1$ values between gases from Block HD4 and gases from its neighboring blocks reaching 10‰. This type of abnormal gas has never been reported previously in the Tarim Basin and such large variations in $\delta^{13}C$ have rarely been observed in other basins globally. Based on a comprehensive analysis of gas geochemical data and the geological setting of the Carboniferous reservoirs in the Hadexun Oilfield, we reveal that the anomalies of the gas compositions and carbon isotope ratios in the Donghe Sandstone reservoir are caused by gas diffusion through the poorly-sealed caprock rather than by pathways such as gas mixing, microorganism degradation, different kerogen types or thermal maturity degrees of source rocks. The documentation of an in-reservoir gas diffusion during the post entrapment process as a major cause for gas geochemical anomalies may offer important insight into exploring natural gas resources in deeply buried sedimentary basins.

1. Introduction

Geochemical characteristics of natural gases have been considered as sensitive tracers for the origin of natural gases and hydrocarbon charging history because their isotopic and compositional signatures contain important information on their genetic and post-genetic histories.

Many scholars considered that 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) including gas sources and degree of thermal evolution are the main factors controlling compositional and isotopic characteristics of hydrocarbon gases. Hydrocarbon gaseous compounds tend to be enriched in ^{13}C as thermal maturity increases. Moreover, most field/laboratory observations and theoretical models of kinetic isotope effects (KIEs) have created a normal sequence of carbon isotopic compositions with $\delta^{13}C$ methane (C_1) < $\delta^{13}C$ ethane (C_2) < $\delta^{13}C$ propane (C_3) and < $\delta^{13}C$ butane (C_4) in natural gas accumulations worldwide (Stahl, 1974; Schoell, 1983, 1988; Chung et al., 1988; Clayton, 1991; Berner et al., 1995; Rooney et al., 1995; Guo et al., 2009; Pan et al.,2010). Apart from genetic elements, some scholars have emphasized the influence of post-genetic elements on compositional and isotopic characteristics, such as migration along the carrier beds (Smith et al., 1971; Prinzhofer and Huc, 1995), gas diffusion (Krooss, 1988; Krooss et al., 1992a, 1992b), adsorption–desorption (Friedrich and Juntgen, 1972), microorganism degradation (James, 1990.; Whiticar, 1994, 1999; Pan et al., 2006; Lu et al., 2010), and gas mixing (Dai et al., 2004; Burruss and Laughrey, 2010). More specifically, gas diffusion from the caprock can separate the original gases into two portions: segregated and residual gases. Compared to the original gases, gaseous hydrocarbons in the segregated gases are enriched in methane but depleted in ^{13}C, and the $\delta^{13}C$ values of individual alkane gases show a normal pattern. However, gaseous hydrocarbons in the residual gases are enriched in C_{2+} compounds (the hydrocarbon gas

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compositions except for methane) since methane diffuses preferentially, and the $\delta^{13}C$ values of individual alkane gases show a reversal pattern (Prinzhofer and Pernaton, 1997; Zhang and Krooss, 2001). Microorganism degradation can also present an isotopic reversal pattern because of preferential consumption of ^{12}C (Coleman et al., 1981; Whiticar, 1999). Chemical oxidation of gaseous compounds, such as that by thermochemical sulfate reduction (TSR), can result in the isotopic reversal of CO_2, rather than in the isotopic reversal of gaseous hydrocarbons (Krouse et al., 1988; Pan et al., 2006; Mankiewicz et al., 2009; Lu et al., 2010). The mixing of gases from different sources or those generated at different levels of thermal maturity can lead to partial isotopic reversal in isotopic compositions (Jenden et al., 1993a,b; Dai et al., 2004; Huang et al., 2004).

In recent years, a type of abnormal gas in the Carboniferous Donghe Sandstone reservoir in the Hadexun Oilfield, Tabei Uplift, Tarim Basin, northwest China has been observed. In comparison with gases from neighboring areas, the gases from the Donghe Sandstone reservoir in Block HD4 in the Hadexun Oilfield are characterized by obvious gas composition and carbon isotope ratio anomalies. Methane tends to become enriched in ^{13}C with the variations in $\delta^{13}C$ values between the gases from Block HD4 and those from its neighboring areas reaching 10‰. This type of abnormal gas has never been reported previously in the Tarim Basin and such large variations in $\delta^{13}C$ are rarely observed in other basins globally. Great geological and geochemical significance arises from the question as to whether the current models of hydrocarbon gas generation and post-genetic elements could rationally explain the composition and carbon isotope ratio anomalies of the gases in the Hadexun Oilfield. In this study, by analyzing data of gas compositions (97 samples) and carbon isotope ratios (17 samples), and integrating these data with the geological setting, we attempt to reveal the cause of the composition and the carbon isotope ratio anomalies of the gases from the Donghe Sandstone reservoir and identify the gas accumulation mechanism. The purpose of this study is to improve the geochemical theory of explaining and interpreting natural gas systems based on data from commercial gas accumulations rather than though laboratory simulation experiments. Moreover, the Hadexun Oilfield is the first significant oilfield discovered in the marine clastics in the Tabei Uplift of the Tarim Basin with proven reserves of over 100 million tons (Zhou et al., 2007). Understanding the cause of the geochemical anomalies and the accumulation mechanisms of the natural gas will shed light on the conditions under which highly productive gas may accumulate and get preserved, and in turn guide further hydrocarbon exploration in the study area.

2. Geological setting

Exploration activities and production of marine hydrocarbons in the Tarim Basin, northwest China, have increased dramatically in recent years (Zhu et al., 2012, 2014). The Hadexun Oilfield, the first significant oilfield discovered in the marine clastics in the basin, has attracted the attention of many scholars. The Hadexun Oilfield lies in the transition zone between the Tabei Uplift and Manjiaer Depression, and bordered by the Lunnan Low Uplift to the north, the Halahatang Sag to the northwest, and the Manjiaer Depression to the south (Fig. 1). The oilfield is divided into three compartments: Block HD4 in the southwestern part, Block HD1 in the southeastern part, and the Northern Block in the northern part (Fig. 1) (Tao et al., 2014). The oilfield is located on a simple structural trap. It is largely a slope structure with few faults. Only the YN1 fault belt occurs in the western part of the Hadexun Oilfield from the interpretation of newly acquired 2D and 3D seismic data, and there is fault-controlled hydrocarbon migration and accumulation in the study area (Fig. 1) (Xu et al., 2008). The target Carboniferous Donghe Sandstone reservoir is distributed widely in the Hadexun Oilfield and is characterized by a low structural profile, large oil-bearing area, thin oil pays, great burial depth, strong reservoir heterogeneity, and complex oil-gas-water distribution. It is thus quite challenging for exploration and development (Zhou et al., 2007; Sun et al., 2011).

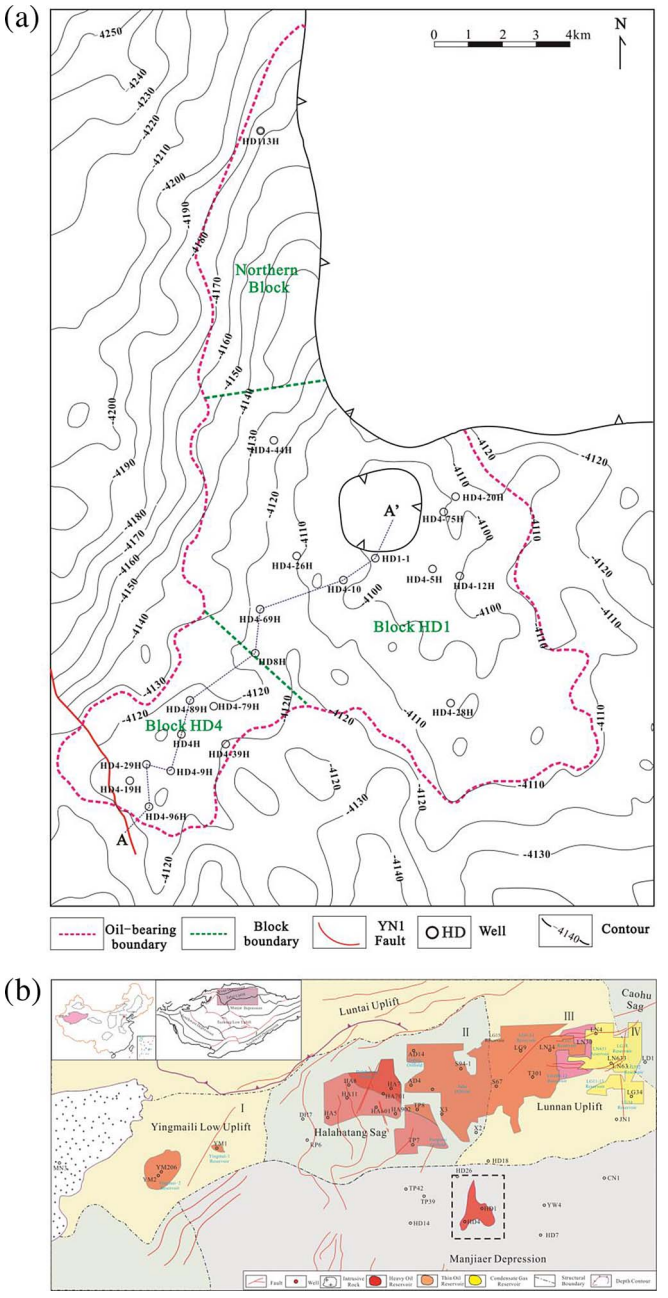

Fig. 1. Map showing the structural elements and the hydrocarbon distribution in the Carboniferous Donghe Sandstone in the Hadexun Oilfield, Tabei Uplift. (a) Map showing the structural elements of the Hadexun Oilfield. The three blocks are as follows: Block HD4 in the southwestern part, Block HD1 in the southeastern part and the Northern Block; (b) map showing the location of the Hadexun Oilfield in the Tabei Uplift of the Tarim Basin, northwest China.

Drilled formations in the Hadexun Oilfield include the Silurian, Carboniferous, Permian, Triassic, Jurassic, Cretaceous, Paleogene, Neogene, and Quaternary, with the Upper Silurian and Devonian missing. From the bottom to top, the Carboniferous consists of the Bachu, Kalashayi, and Xiaohaizi Formations. There are two reservoir-caprock assemblages in the Carboniferous (Fig. 2): (1) the Donghe Sandstone in the Bachu Formation as a reservoir and the Bachu Formation breccia as a caprock, and (2) the mudstone in the middle Kalashayi Formation as a reservoir with the limestone in the lower Kalashayi Formation and mudstone in the upper-Kalashayi Formation as