



Predicting the proportion of free and adsorbed gas by isotopic geochemical data: A case study from lower Permian shale in the southern North China basin (SNCB)



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ABSTRACT

The alternating marine-terrestrial shale facies of the Lower Permian Taiyuan (P_{1t}) formation and the Lower Permian Shanxi (P_{1s}) formation in the north margin of the southern North China basin (SNCB) are characterized by their high TOC values (1.76–5.09%), types II and III organic matter, and high Ro values (>3.0%). Geochemical parameters of 12 gas samples from the Lower Permian shale formations from well Weican-1 were analyzed in this study. The gases are dominated by methane, with small amounts of ethane, without propane and butane. Wetness of the gas is only 0.25–0.58% reflecting extremely high maturity of the source rock. The $\delta^{13}\text{C}_1$ values range from -31.6‰ to -26.8‰ and the $\delta^{13}\text{C}_2$ values range from -35.9‰ to -33.2‰ , the $\delta^2\text{H}_{\text{CH}_4}$ values range from -221.1‰ to -187‰ . Furthermore, carbon isotopic compositions of the alkane gases from the Lower Permian shale are characterized by $\delta^{13}\text{C}_1 > \delta^{13}\text{C}_2$, this indicates that the gases released from Permian shale are of thermogenic origin and mostly sourced from the continental shale and coal measures, with minor contribution from oil cracked gas from marine mudstones. Geochemical fractionation during the adsorption/desorption process of the shale system may play a significant part in influencing $\delta^{13}\text{C}_1$ values of shale gas. The results show that the $\delta^{13}\text{C}_1$ becomes heavier with increasing degree of gas desorption. Based on isotope fractionation during desorption of gas in shales, an equation was established to estimate the proportion of free and adsorbed gas in shales using $\delta^{13}\text{C}_1$ of shale gas. In comparison with other equations, this equation is based on the direct data of gas desorption experiment to avoid the adsorbed gas content often exhibit maxima from excess sorption isotherms experiment. This method provides efficient way to understand the gas storage behavior in shales and broaden the application of gas isotope.

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1. Introduction

Shale gas is one of the most important unconventional hydrocarbon resources in which natural gas is found as adsorbed gas within organic matter and on inorganic minerals, free gas within fractures and intergranular porosity, and as well as dissolved gas in kerogen, oil and water (Schettler and Parmely, 1990; Martini et al., 1998). Adsorbed gas may account for 20–85% of total gas in shale reservoirs (Hill and Nelson, 2000). Thus unlike conventional gas accumulations, in which gas is present mainly as free gas, or coalbed methane accumulations, where gas is considered to be mainly adsorbed, both of free gas and adsorbed gas may attribute to gas production from shales.

To understand gas occurrence and storage mechanism in shales is a key question for shale gas reservoir engineering. One commonly used method is to use reservoir simulation for representation, however, this

will require a reservoir simulator that faithfully represents the shale gas storage and flow behaviors. Moreover, because adsorbed gas may take a longer time to transport through the matrix than free gas (e.g. Yuan et al., 2014), the contribution of adsorbed and free gas may be even more complex.

Different stages of basin evolution directly control the development and distribution of organic-rich shale (Li et al., 2009). According to the depositional environment, organic-rich shale can be divided into marine shale, marine-terrestrial coal bed carbonaceous shale and continental lacustrine derived shales (Zou et al., 2010). Coal bed shale (shale derived from mixed marine-terrestrial sources) was formed in marine-terrestrial transitional deltaic facies, such as the Carboniferous Benxi Formation and the Lower Permian Shanxi Formation–Taiyuan Formation in Ordos Basin, the Carboniferous–Permian in Junggar Basin, the Carboniferous–Permian in Tarim Basin, the Carboniferous–Permian in Northern China, and the Permian Longtan Formation in Southern China. It can also be a major source rock for large-scale oil/gas fields. For example, the Upper Paleozoic carbonaceous shale in Ordos Basin is the major gas source rock for Sulige and other large gas fields and its

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environment of deposition was deltaic facies. The Triassic–Jurassic and Tertiary developed several sets of carbonaceous shale, which were associated with coal measures of deltaic facies (Zou et al., 2010). Since the oil and gas exploration in the southern North China basin, previous studies confirmed that the coal bed shale from the Lower Permian which formed in alternating marine–terrestrial deltaic facies are widely distributed in this area. But shale gas exploration in this region has not achieved commercial progress due to complex geological conditions such as structural deformation in the southern North China basin. In 2014, the vertical drills (well Weican-1 and Mouye-1) in the Permian shale firstly found shale gas in this region, which marks a major breakthrough in the exploration of alternating marine–terrestrial deltaic facies shale gas in north China.

Natural gas from marine shale (shale gas derived from marine organic matter) has recently gained significant success in the USA, and it triggered a worldwide fever for shale hydrocarbon resources (Curtis, 2002; Tang et al., 2014a, 2014b). In contrast, there has been little work done and less attention paid to alternating marine–terrestrial deltaic facies shale. In this paper, we present stable carbon and hydrogen isotopes of alkane gases as a means to understand the geochemical characteristics of gases from the lower Permian alternating marine–terrestrial deltaic facies shale in the north margin of the southern North China basin. In addition, the generation of shale gas follows a mechanism that is similar to that of conventional gas, which means the origins of shale gas can also be biogenic, thermogenic or a mixture of the two. In conventional gas reservoirs, the variation of gas isotope composition is mainly determined by kerogen isotope composition, maturity and charging efficiency. For shale gas, residual oil in shale may convert to gas and water may involve in this conversion. Products from different chemical processes make the isotope composition deviate from conventional models. Therefore, the reasons for the geochemical particularities of shale gas may be mainly related to the storage stage. We have studied dynamic changes in the geochemical characteristics of shale gas (on the basis of the process of desorption) in order to improve our understanding on the isotope variability of shale gas. Moreover, a simple two-end member mass balance model was proposed to estimate the relative proportion of free gas and adsorbed gas in desorbed gas by using carbon isotope of methane in different stages of the desorption experiment.

2. Geological setting

The southern North China basin (SNCB) is one of the Meso–Cenozoic superimposed basin which is located in the south of North China platform. It is on the western side of the Tan–Lu fault, the northern side of the Qinling–Dabie orogenic belt and the eastern side of the western Henan uplift zone (Lin et al., 2011). The tectonic evolution of this basin is controlled by the Qinling–Dabie orogenic belt and the Tan–Lu fault zone and thus showing the NW–WNNW tectonic pattern. From the north to the south, the southern North China basin can be divided into five secondary tectonic units: the Kaifeng depression, Taikang uplift, Zhoukou depression, Changshan uplift and Xinyang–Hefei depression (Fig. 1). In addition, the secondary tectonic units in the basin can be divided into some episodes of subsidence and uplift, the total area of the basin is 150000 km² (Diao et al., 2011; Lin et al., 2011).

The SNCB is a part of the North China Craton Basin formed during the late Paleozoic. Affected by the sea level rise in Late Carboniferous, the study area began to accept marine derived sedimentation from a transgression, and the evolution process of the epicontinental sea to the continental basin was started (Diao et al., 2011). The depositional process was controlled by regional tectonic movement, and can be divided into three processes. First, the North China platform re-subsided upon entering the Late Carboniferous, and seawater intruded from the northeast to the southwest under the paleotopography background (i.e., high in the south and low in the north), forming the vast North China epicontinental sea basin. The Taiyuan formation, which consists of black shale,

coal, limestone and quartzose sandstone, was deposited in this environment during the early Permian, with a thickness ranging from 30 to 175 m. Second, the collision between the North China plate and the Siberia plate caused largescale tectonic movement and a marine regression from north to south. The river-dominated shallow water delta, which was transitioned from an epicontinental sea via marine regression, was a major depositional environment during the early Permian. The Shanxi formation, which consists of shale, coal, quartzose sandstone, was deposited in this environment with thicknesses ranging from 50 to 130 m. Finally, the North China plate was again uplifted by squeezing action during the late Permian, causing seawater to fully withdraw from the North China platform, forming terrestrial deposition environment (Diao et al., 2011; Duan et al., 2002; Liu et al., 1999; Yu et al., 2005; Zhou et al., 2010).

The organic matter of the two set of source rocks are type II to type III kerogen and dominated by type III kerogen (Dai, 2000; Dang et al., 2016). The TOC of the black mudstones from P₁t and P₁s coal-bearing measures ranges from 0.92% to 4.24% and from 0.44% to 5.10%, respectively (Dang et al., 2016). The maturity of the source rocks in the east and south of the basin is relatively low, the vitrinite reflectance (R_o) values range from 0.5% to 1.0% and from 0.7% to 1.2%, respectively. However, the source rocks in the north of the basin are in the over mature stage with the vitrinite reflectance values over 3.0% (Sun et al., 2014).

3. Samples and methods

In this study, four shale core samples from a depth of 2730–2762 m (WC-1, WC-2, WC-4, WC-6) investigated here were collected from shale gas well Weican-1 in the north margin of the SNCB. Among them, sample WC-1 (2730 m) and WC-2 (2731 m) were from Shanxi formation, and sample WC-4 (2758 m) and WC-6 (2762 m) from Taiyuan formation. The samples are of 10 cm diameter, 4.5 kg, 4.5 kg, 2.97 kg and 4.21 kg in weight respectively. The TOC values of the four shale core samples were 2.27%, 2.78%, 3.62%, 2.15% respectively, and the R_o were 3.2%, 3.2%, 3.5%, 3.7% respectively. The high vitrinite reflectance values (R_o) presented here are also supported by previous studies, which reported that abnormally high thermal maturities (>3.0%) occur in this area, mainly caused by thermal events (Cheng et al., 2011; Wu et al., 2015; Xu et al., 2005, 2011; Zhao et al., 2011). Twelve gas samples were collected from the desorption of the above four core samples, and we collected gas samples at 20 °C, 90 °C and 120 °C for the gas releasing process of each core sample. The molecular composition of gas was performed at the Key Laboratory of Marine Reservoir Evolution and Hydrocarbon Abundance Mechanism, China University of Geosciences, Beijing. The stable carbon and hydrogen isotopes analyses were performed at the Nuclear Industry Beijing Geological Research Analysis and Test Research Centre (<http://www.briug.cn/>).

Desorbed gas content was quantified after the core sample recovered at the drilling site. It was placed as quickly as possible inside a hermetically sealed canister using quartz sand fill gaps and the volumes of gas released inside the canister were periodically measured using a graduated cylinder at atmospheric pressure (Mavor and Nelson, 1997; Ma et al., 2015). The gas releasing process can be divided into three stages (Fig. 2), the amounts of gas released at room temperature of 20 °C were small (stage 1). However, the amounts of gas released at reservoir temperature of 90 °C rapidly increased and the final stabilized (stage 2). A high temperature of 120 °C was used to expedite the adsorbed gas completely released (stage 3). Gas content of each stage determination finished when the released gas fell below 5 ml in 2 h.

The molecular composition of the gas samples was determined by Agilent 6890 N gas chromatograph (GC) equipped with a flame ionization detector and a thermal conductivity detector. Individual hydrocarbon gas components (C₁–C₂) were separated using a capillary column (PLOT Al₂O₃ 50 m × 0.53 mm). Nonhydrocarbon gases were separated using two capillary columns (PLOT Molsieve 5 Å 30 m × 0.53 mm, PLOT Q 30 m × 0.53 mm). The GC oven temperature was initially set

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