



Original research paper

Variation in the carbon isotopic composition of alkanes during shale gas desorption process and its geological significance[☆]

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Abstract

Taking Chang 7 shale of the Yanchang Formation in the southeastern area of the Yishan slope of the Ordos Basin as our research subject, desorption experiments were performed on more than 30 shale samples that helped this study focus on the variation of carbon isotopic composition of alkanes during the desorption process of shale gas, the possible causes of this phenomenon and its geological significance will also be discussed as follows. It was found that carbon isotopic composition became higher by 9.2‰ (from −50.1‰ to −40.9‰) for methane desorbed gas ($\delta^{13}\text{C}_1$), and it also became higher by 2.8‰ (from −35.5‰ to −32.7‰) for ethane ($\delta^{13}\text{C}_2$), but there's barely any differences for propane during the desorption process. At room temperature or constant temperature, carbon isotopic composition for both methane and ethane ($\delta^{13}\text{C}_1$ & $\delta^{13}\text{C}_2$) increased continuously. The values of $\delta^{13}\text{C}_1$ and $\delta^{13}\text{C}_2$ were low at first and then it gradually became high whilst the desorption process of shale gas that was optimized with the aid of the increasing temperature. The reason for this phenomenon is most likely due to the main adsorption/desorption of shale gas and diffusion migration fractionation of isotope. Additionally, these variation characteristics of methane and ethane carbon isotope composition of shale gas may be a vital reason for the relatively low carbon isotopic composition of methane compared to $\delta^{13}\text{C}$ calculated by means of the vitrinite reflectance (R_o), and this may be applied to evaluate the remaining amount of shale gas resources. Copyright © 2016, Lanzhou Literature and Information Center, Chinese Academy of Sciences AND Langfang Branch of Research Institute of Petroleum Exploration and Development, PetroChina. Publishing services by Elsevier B.V. on behalf of KeAi Communications Co. Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

Keywords: Shale gas; Carbon isotopic composition; Desorption; Chang 7 shale of Yanchang Formation; Geological significance

1. Introduction

Carbon isotopic composition of alkanes is one of the most important indicators in identifying the origin of natural gas and correlating gas sources [1–4]. Based on the relationship

between measured carbon isotopic composition of methane ($\delta^{13}\text{C}_1$) and the vitrinite reflectance of source rock (R_o), various models have been built to estimate the thermal maturity of source rocks based on the $\delta^{13}\text{C}$ of associated natural gas [2,3,5–7]. However, recent research shows that the geochemical characteristics of shale gas have a certain particularity compared to the conventional natural gases, not to mention, appropriately applied geochemical theory to conventional natural gas is not suitable for shale gas.

Shale gas retains and accumulates natural gas in source rocks and it's characterized as self-generating and self-storing. Shale gas is mainly stored in shale reservoirs as both free and

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adsorption gas; the proportion of adsorption gas ranges from 20% to 85% (average 50%) of the total gas in shale [8–10]. It is recognized that adsorption gas is largely part of the shale gas which has a close relationship with the gas content in the shale. At present, It has been comprehended that the process of adsorption and desorption was a key factor in controlling the occurrence and development of shale gas [11]. So far there's only a minimal amount of research that's focused on the variation of carbon isotopic composition of alkanes during the shale gas desorption. Gao et al. [12] studied the characteristics of separated shale gas from the upper Triassic of the Junggar Basin, but it only partly reflects the variation process of carbon isotopic composition of alkanes during the desorption due to brief desorption time and limited samples.

Ordos Basin is the second largest sedimentary basin in China, it is rich in oil, gas, and coal resources; the basin basically developed multi-effective hydrocarbon source rocks. The Chang 7 lacustrine dark shale of the Yanchang Formation is the main source rock of the Ordos Basin in Mesozoic. The Chang 7 lacustrine dark shale is abundant in organic matters and it's in the mature stage $R_o > 1.0\%$ for most areas of the basin; the "L-type" area of Dingbian-Huachi-Fuxian may be the most developed area for shale gas [13]. The southeastern area of the Yishan slope of the Ordos Basin is located in the center of the lacustrine basin in Mesozoic. The Chang 7 shale is widely distributed in this area, and it is characterized by being extremely thick, having moderate burial, it possesses a high TOC value, and it has a rich shale gas resources [14–16]. The shale core samples were collected from the Chang 7 shale of the Yanchang Formation in the southeastern area of the Yishan slope of the Ordos Basin and it was used in field desorption experiments for this paper. The variation of carbon isotopic composition of alkanes during the shale gas desorption and the possible reasons for this phenomenon were studied in this paper. We made a study of particular geochemical characteristics of shale gas that may help us further understand the formation, storage, and accumulation mechanism of shale gas.

2. Material and methods

Shale core samples were collected from two new shale gas wells of the Triassic Yanchang Formation Chang 7 member in the southeastern Yishan slope. The sampling position was located in the central zone of the lacustrine shale deposits in the Yanchang Formation. Shale thickness was approximately 70 m within the sampling area, sampling depth ranged from 1290 m to 1365 m, TOC values went up to 4%–5%, and R_o was 1.0%. The samples were 10 cm diameter and were about 4–5 cm long. The shale core samples that were extracted were immediately placed in a transparent sealed canister that was filled with water and upside and then the cap of the canister was covered. Gas was witnessed to be given off from the rock into the canister. Gas desorption increased the pressure in the canister and caused a portion of the water to be expelled out through the gap in the bottom of the canister. After a month, the core sample was removed from the canister and is placed

into a new canister. The desorbed gas was collected in a glass bottle by initially discharging water, this served as the first gas sample (A). Its volume was measured and the material was tested for its chemical and isotopic composition. The core sample continued to desorb through heating in the new canister, the heating temperature was 80–90 °C. The temperature remained constant for 28.5 h and the desorbed gas was collected in another glass bottle, this serves as the second gas sample (B). We also randomly selected some shale core samples that were used in the desorption experiment that vary the previous procedure; desorption experiment could be divided into 17 stages. By repeating the desorption experiment on the same set of shale samples in different temperature conditions, the gas could be considered fully released from the shale samples, exclusive the amount of residual gas. The desorbed gas in the glass bottle was collected according to a particular time interval. Its volume was measured and the material was tested for its chemical and isotopic composition. The variation of carbon isotopic composition of alkanes of desorbed gas was observed. This method can reflect the dynamic changes of carbon isotopic composition of alkanes during the shale gas desorption process. The experimental analysis was performed in the Key Laboratory of Petroleum Resources Research, Chinese Academy of Sciences (Lanzhou).

3. Results

3.1. The chemical compositions variation of desorbed gas

The analytical results of hydrocarbon gas composition of the desorbed gas show that the dryness coefficient (C_1/C_{1-5}) of desorbed gas had significant changes. The desorbed gas at room temperature (i.e. early desorbed gas) is dryer than the heated one (i.e. late desorbed gas), and the dryness coefficient of desorbed gas decreases with the increasing depth. The dryness coefficient of early and late desorbed gas ranges from

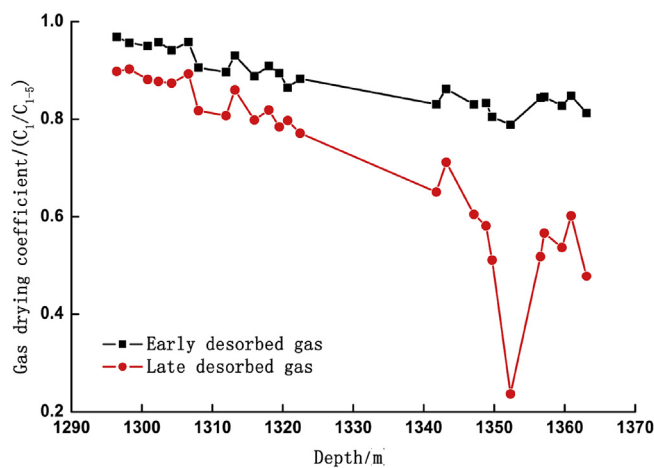


Fig. 1. The wetness (C_1/C_{1-5}) variation of desorbed gas from Chang 7 shale of the Yanchang Formation.

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