Geochemical characteristics and genesis of the Upper Paleozoic natural gas in Longdong area, Ordos Basin, NW China

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Abstract. The Upper Paleozoic gases from the Longdong areas in the Ordos Basin are characterized by low contents of heavy hydrocarbon gases, the mean value of wetness as 2%, relatively heavy δ¹³C₁(δ¹³C₁ value -28.3‰), and large variations in δ¹³C₂⁺. The distribution pattern of carbon isotopic sequence of natural gas is mainly characterized by the abnormal light δ¹³C₂⁺ values and carbon isotopic reversal. According to the organic geochemical characteristics of natural gas and the development of gas-source rocks, the upper Paleozoic natural gas in Longdong area belongs to high mature and over-mature "coal-formed gas". In theory, only the mixture of highly mature gas with extremely low contents of heavy hydrocarbon gases and a small amount of natural gas with higher contents and lighter carbon isotopic composition of heavy hydrocarbon gases can lead to the occurrence of abnormally light δ¹³C₂⁺ and the carbon isotope reversal between methane and ethane. The mixed gas is probably derived from the thermal cracking of a small amount of residual soluble organic matter in the gas-source rocks and reservoirs during the initial stage of the uplift movement at the end of Early Cretaceous.

Key words: Ethane carbon isotope; Inverted sequence of carbon isotopic composition; Natural gas of high maturity; natural gas genesis; Ordos Basin.

1. Introduction

The Ordos Basin is a large-scale multicycle craton basin with simple structure, which covers Shaanxi, Gansu, Ningxia, Inner Mongolia and Shanxi provinces, with an area of about 25 × 10⁴ km². Longdong area is located in the southwest of the Ordos Basin, spanning the southwest of Yishan slope and the southern end of Tianhuan depression. It is a composite oil and gas accumulation area of Mesozoic middle-shallow reservoirs and Paleozoic deep gas reservoirs, with an area of about 4.5 × 10⁴ km² (Fig. 1).

After nearly 50 years of exploration, along with the deepening understanding of the geological laws of hydrocarbon generation and accumulation and the continuous progress of exploration and development technology, the oil and gas exploration objects in Longdong area have gradually expanded
from Mesozoic middle-shallow reservoirs to Paleozoic deep gas reservoirs, and experienced three stages of exploration and development: petroleum exploration (before 2000), regional exploration of natural gas (2000-2012) and Integration of exploration and development (2013-2017). The established geological reserves of the member 8 of Shihezi Formation (P2h8) and the member 1 of Shanxi Formation (P1s1) of the Upper Paleozoic Permian, are more than 2000 × 10^8 m³, and the proved reserves of P1s1 gas-rich block are 318.86 × 10^8 m³, which made Qingyang gas field discovered (Fu et al., 2019). The average buried depth of the central Qingyang gasfield is 4276.0m, so it belongs to large deep gas field. The stratigraphic division of the Upper Paleozoic in Longdong area is shown in Table 1.

Fig. 1 Sketched map showing the location of Longdong area in the Ordos Basin

2. Geochemical characteristics of natural gas

2.1 Characteristics of natural gas components
Natural gas in Longdong area of the Ordos Basin is mainly produced from tight sandstone reservoirs of P2h8 and P1s1 of Upper Paleozoic. Natural gas chromatographic data reveal that the upper Paleozoic natural gas components in Longdong area are similar to Sulige gas field in the Ordos Basin (Table 2). They are mainly both hydrocarbon gas, with hydrocarbon component content more than 95%, Non-hydrocarbon gases are mainly CO₂ and N₂, and well ZT1 and well ZT2 contain trace hydrogen component; the content of CH₄ in hydrocarbon gas is absolutely dominant, the content of C₂+ heavy hydrocarbon component is low, and C₁ / ΣCₙ ratio is > 0.95, with an average of 0.98, which shows typical "dry gas" characteristics.

2.2 Carbon isotope characteristics of natural gas components
According to the carbon isotope test results of natural gas components (Table 3), the δ¹³C₁ values of natural gas from P2h8, P1s1 and P1s2 (the member 2 of Shanxi Formation of the Upper Paleozoic Permian) in Longdong area are distributed between −30.1 ‰ and −24.1 ‰, with an average value of −28.3 ‰; The distribution range of δ¹³C₃ is relatively wider, ranging from −36.1 ‰ to −23.0 ‰, with an average value of −30.3 ‰. Among the 10 tested data, the δ¹³C₃ value of 7 samples is < −28.5 ‰, accounting for 70% of the statistical samples; The δ¹³C₅ value was significantly lighter, mainly distributed in −32.1 ‰ to −25.5 ‰, with the average of −31.4 ‰, and 90% of the tested samples were < −26.5 ‰.

Compared with other blocks in the basin (Table 3), the carbon isotopic characteristics of the upper Paleozoic natural gas components in Longdong area are similar to those in southern Sulige, Gaoqiao and Yichuan- Huanglong areas, where the regional thermal evolution degree is also over mature, showing the characteristics of heavier δ¹³C₁ value but significantly lighter δ¹³C value of C₂+ heavy hydrocarbon component. In the northeastern part of the basin and most of Sulige area with the relatively
low regional thermal evolution degree. The carbon isotopic of the upper Paleozoic natural gas component is generally heavier, the average of $\delta^{13}$C$_1$, $\delta^{13}$C$_2$ and $\delta^{13}$C$_3$ values are −30.2‰, −24.54‰ and −24.4‰, respectively.

Table 1. The Paleozoic stratigraphic division of Longdong area in the Ordos Basin

| Strata                | Lithology                              |
|-----------------------|----------------------------------------|
| Erathem               |                                        |
| Upper Permian         | Calcareous nodule mudstone, sandy mudstone and flesh-red sandstone. |
| Middle Permian        | Purple-red and yellow-green mudstone, sandy mudstone and ligh-gray coarse sandstone. |
| Lower Permian         | Dark gray and gray black sandy mudstone, imbedded with coal seam. |
| Paleozoic             |                                        |
| Lower Paleozoic       | Black mudstone imbedded with limestone and coal seam. |
| Upper Paleozoic       | Dark gray, brown gray residual granular dolomite and oolitic dolomite, massive limestone. |
| Odovician             |                                        |
| Lower Odovician       | Carbonate                              |
| Cambrian              |                                        |
| Middle Cambrian       |                                      |

2.3 Carbon isotope series distribution model of alkane gas components

The carbon isotopic series fractionation model of alkane gas components is an important indicator for distinguishing the characteristics and genetic types of natural gas. The regular carbon isotopic series ($\delta^{13}$C$_1 < \delta^{13}$C$_2 < \delta^{13}$C$_3$) is a sign of primary alkane gas of organic origin. In the upper Paleozoic of the Ordos Basin, this type of natural gas is mainly distributed in the northeast of the basin with relatively low regional thermal evolution degree (Fig. 2a), where $\delta^{13}$C$_1-\delta^{13}$C$_2-\delta^{13}$C$_3$ show inclined "regular L" shape on the fractionation model diagram. The carbon isotope series of alkane gas components in most areas of Sulige are basically the shape of "V", and partially reversed by $\delta^{13}$C$_1 < \delta^{13}$C$_2 > \delta^{13}$C$_3$ (Fig. 2b); The carbon isotope series of alkane gas components in the Upper Paleozoic reservoirs in Longdong area are mainly characterized by complete inversion of $\delta^{13}$C$_1 > \delta^{13}$C$_2 > \delta^{13}$C$_3$ or partial inversion of $\delta^{13}$C$_1 > \delta^{13}$C$_2 < \delta^{13}$C$_3$, so that $^{13}$C$_1-\delta^{13}$C$_2-\delta^{13}$C$_3$ show inverted "V" shape (Fig. 2c), and individual samples showed partial inversion of $\delta^{13}$C$_1 < \delta^{13}$C$_2 > \delta^{13}$C$_3$.

Fig. 2 Carbon isotope distribution patterns of natural gas alkanes in Upper Paleozoic from different areas of the Ordos Basin
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been used as the main indic
 evolution degree of organic matter, natural gas migration and some secondary changes, so it has long
important basis for distinguishing the genetic types of natural gas. The carbon
isotope fractionation model of the
Sulige area in Ordos basin is relatively higher (δ13C = 2.8‰), the thermal evolution degree of organic matter in
Upper Paleozoic strata has reached over-mature stage (R0 > 2.0%).

3. Genetic analysis of natural gas

3.1 Distinguishing the genetic types of natural gas
The carbon isotope characteristics of natural gas components and alkane gas components are the
important basis for distinguishing the genetic types of natural gas. The carbon isotopic composition of
heavy hydrocarbon components such as ethane is relatively stable, which is less affected by the thermal
evolution degree of organic matter, natural gas migration and some secondary changes, so it has long
been used as the main indicator for identifying genetic types of natural gas (Zhang, 1994; Chen, 1994;
Huang et al., 1996). In recent years, with the target strata and research objects of natural gas exploration
have extended from middle-shallow layer to deep and ultra-deep layer, from conventional to
unconventional, the carbon isotopic composition of natural gas components shows different "abnormal"
phenomenon that the carbon isotopic composition of heavy hydrocarbon components such as ethane and
propane is significantly lighter and the carbon isotopic series of alkane gas components is partially or
completely reversed, which not only increases the difficulty in identifying the genetic types of natural
gas, but also makes the understanding of carbon isotope dynamic fractionation mechanism in the process
of natural gas generation faced more challenges.

According to the analysis of the geochemical characteristics of natural gas, the C1 / ΣCn ratio of the
upper Paleozoic P2h8 and P1s1 in Longdong area is > 0.95, with an average of 0.98, and the average of
δ13C1, δ13C2 and δ13C3 values are -28.3‰, -30.3‰ and -31.4‰, respectively. Therefore, it is specified
that this kind of natural gas belongs to organic genetic dry gas of high thermal evolution. The δ13C1 of

Table 2. Comparison of the Upper Paleozoic natural gas components between Longdong area and
Sulige area in the Ordos Basin

| Exploration zone | Well | Strata | Natural gas components (%) | C1/ΣCn |
|------------------|------|--------|-----------------------------|--------|
| Longdong         | HT2  | P2h8   | 96.309 1.102 0.134 0.021 0.025 2.221 0.162 0.987 |
|                  | L1   | P1s1   | 93.142 0.788 0.184 0.010 0.012 4.339 1.506 0.989 |
|                  | L11  | P2h8   | 93.838 0.653 0.135 0.007 0.005 2.718 2.642 0.992 |
|                  | QT3  | P1s1   | 92.408 1.113 0.131 0.014 0.017 5.272 1.030 0.986 |
|                  | ZT1  | P1s1   | 94.610 1.343 0.127 0.013 0.012 0.000 2.316 0.984 |
|                  | ZT2  | P1s1   | 93.660 1.952 0.447 0.093 0.089 2.086 1.448 0.972 |
|                  | T40  | P2h8   | 89.426 2.059 0.298 0.021 0.044 7.712 0.416 0.973 |
| Sulige           | T40  | P1s1   | 93.551 3.018 0.443 0.061 0.062 1.985 0.812 0.962 |
|                  | S368 | P2h8   | 91.876 0.953 0.139 0.012 0.016 4.717 2.263 0.988 |
|                  | S112 | P2h8   | 94.031 1.870 0.283 0.032 0.037 2.999 0.720 0.977 |
|                  | S112 | P1s1   | 95.387 1.808 0.013 0.190 0.037 2.175 0.352 0.979 |
|                  | S143 | P1s1   | 97.305 2.055 0.287 0.044 0.035 0.000 0.183 0.975 |
the upper Paleozoic natural gas in Longdong area is relatively heavier, which demonstrates the characteristics of high and over mature "coal-formed gas", while the carbon isotopic composition of heavy hydrocarbon components such as ethane is significantly lighter, and the distribution patterns of carbon isotope series of natural gas components mainly show the inverted "V" shape of δ³⁴C₁ > δ³¹C₂ < δ³¹C₃ or the right upward linear distribution of δ³¹C₁ > δ³¹C₂ > δ³¹C₃, and the average of δ³¹C₂ minus δ³¹C₁ is 3.04 ‰.

| Exploration zone | Well | Strata          | δ¹³C(PDB) % | C₁   | C₂   | C₃   |
|------------------|------|----------------|-------------|------|------|------|
|                  | CT2  | P₂h₈s          | -28.4       | -32.2| -32.1|
|                  | L4   | P₁s₁           | -28.8       | -23.0| -25.5|
|                  | L7   | P₂h₈s, P₁s₁    | -25.9       | -33.6| -32.9|
|                  | L20  | P₃s₂           | -29.6       | -29.1| -30.2|
|                  | L22  | P₃s₂           | -30.1       | -36.1| -35.6|
|                  | QS2  | P₂h₈s          | -27.6       | -26.8| -31.9|
|                  | CT3  | P₂h₈s          | -30.4       | -31.9| -33.0|
|                  | QT1  | P₁s₁           | -29.5       | -29.8| -30.7|
|                  | L5   | P₁s₁           | -27.4       | -33.4| -31.8|
|                  | Q1-13-65 | P₁s₁       | -24.1       | -27.5| -30.3|
|                  | S371 | P₂h₈s, P₁s₁    | -32.45      | -34.28| -30.13|
|                  | S127 | P₂h₈s          | -28.99      | -33.7| -34.06|
|                  | LN32 | P₂h₈s          | -32.30      | -30.30| -30.46|
|                  | S316 | P₁s₂, C₂b      | -29.11      | -36.43| -35.20|
|                  | S339 | P₁s₂           | -29.06      | -34.20| /    |
|                  | Y7   | P₂t            | -35.14      | -36.18| -33.7|
|                  | Y8   | P₁s₂           | -31.28      | -31.39| -30.84|
|                  | M10  | P₃q₈s          | -34.47      | -27.46| -24.4|
|                  | M17  | P₂h₈s          | -34.88      | -25.55| -23.17|
|                  | SH23 | P₂t            | -35.71      | -23.67| -22.63|
|                  | SH34 | P₂h₈s          | -37.22      | -27.48| -26.18|
|                  | T46  | P₁s₁           | -33.81      | -22.99| -22.58|
|                  | S353 | P₁s₁, P₂h₈s   | -24.08      | -25.57| -28.7 |
|                  | S388 | P₁s₁           | -28.76      | -23.21| -25.13|
|                  | S120 | P₁s₁           | -28.30      | -21.71| -22.30|
|                  | S134 | P₂h₈s          | -32.49      | -23.61| -25.32|

When studying the genetic types of Lower Paleozoic Ordovician Natural Gas in the west Jingbian of the Ordos Basin (Kong et al., 2016), the author analyzed the relationship of the ΣC₂⁺ versus δ³¹C₂ and C₁ / ΣCn versus δ³¹C₂ of Paleozoic natural gas in the whole basin. The results show that when C₁ / ΣCn value is > 98% and ΣC₂⁺ content is < 2%, δ³¹C₂ value decreases significantly with the increase of methane content and decrease of heavy hydrocarbon content. It can be seen that the carbon isotopic composition of heavy hydrocarbon components such as ethane is not stable for high and over mature natural gas so as not effectively to reflect the source of original parent material. Although the dynamic fractionation of carbon isotopic composition of methane is relatively significant, and the carbon isotope composition difference between "oil-formed gas" and "coal-formed gas" decreases after entering the high thermal evolution stage, it should still be used as the main indicator for the genetic determination.
of natural gas (Yang et al., 2009; Kong et al., 2016). Significantly lighter as the carbon isotopic composition of heavy hydrocarbon components such as ethane of the upper Paleozoic natural gas in Longdong area is, the δ13C value is similar to the typical "coal-formed gas" of the upper Paleozoic in Sulige area, obviously heavier than the oil-type gas of self generation and self accumulation of the Lower Paleozoic Ordovician subsalt. Thus, the upper Paleozoic natural gas in Longdong area belongs to high and over mature "coal-formed gas" (Fig. 3).

In addition, from the perspective of the accumulation characteristics, the Upper Paleozoic Carboniferous-Permian in the Longdong area developed a set of alternate marine and continental coal-bearing formation with the main source rock types of thin coal seam and dark mudstone, which are mainly distributed in Taiyuan Formation of Lower Permian(P1t) and P1s2. The thickness of coal seam is thin and unevenly distributed that the east and west sides are relatively large(2~5m), while the south, north and central areas are thin(< 2m, even lacuna in some areas). The thickness of dark mudstone is 30m ~ 80m, which is overally thick in the north but thin in the south. The Upper Paleozoic coal rock in the area belongs to humic coal, whose macerals are mainly vitrinite and inertinite with a small amount of exinite. The types of kerogen of dark mudstone are mainly humic and mixed II, with the δ13C value of kerogen ~23.36%, which reflects that the biological input of dark mudstone is mainly composed of higher plants. The TOC of the Upper Paleozoic coal in Longdong area is in the range of 40.23% – 91.32%, with the TOC value of dark mudstone 5.28% and 3.15% respectively in P1t and P1s2 and high thermal evolution degree of gas source rock(Ro is in the range of 2.17%~3.02%), which can provide sufficient gas source for tight sandstone reservoir of Upper Paleozoic, thus forming in-source and near-source gas reservoir. Longdong area , located in the central paleouplift of Ordos Basin, due to the uplift of the central paleouplift and the long-term exposure of Ordovician carbonate strata, was exposed to certain denudation so that the Ordovician strata are thin and the marine source rocks are underdeveloped.

3.2 Analysis of negative carbon isotope composition of ethane and other heavy hydrocarbon components
In the past, the reversal of the carbon isotope sequence of alkane gas components in that the carbon isotope composition of heavy hydrocarbon components such as ethane was significantly lighter was mostly attributed to various mixed-source effects or secondary effects such as bacterial oxidation(Dai et al., 2004; Burrus et al., 2010), though the mixed oil-formed gas lower Paleozoic in the Ordos Basin can't make the carbon isotope composition of heavy hydrocarbon components such as natural gas ethane and propane significantly lighter (Kong et al., 2016), so it is necessary to conduct further analysis for its genesis.

From the perspective of the generation and thermal evolution process of gaseous hydrocarbons, methane is the final product of thermal degradation and cracking of organic parent materials and hydrocarbons, while heavy hydrocarbon components such as ethane are a kind of intermediate products which will be cracked at higher temperatures(Zhang et al., 2016). In the process of organic matter thermal evolution, the generation and evolution of heavy hydrocarbon components such as ethane in natural gas can be classified into two stages: generation of thermal degradation of organic parent material (the whole thermal evolution stage) and the thermal cracking of liquid hydrocarbon (high over mature stage), cracking and extinction of itself (Zhang et al., 2016). Theoretically, the carbon isotope composition of heavy hydrocarbon components such as ethane will become heavier in these processes with the improvement of thermal evolution degree. However, it is very likely that heavier carbon isotope composition of methane and high mature natural gas with extremely low heavy hydrocarbon content such as ethane, mixed with soluble organic matter cracked gas with high concentrations of ethane and lighter carbon isotopic composition of gas components, are responsible for which the carbon isotopic composition of heavy hydrocarbon components such as ethane is lighter.

Longdong area is located in the southwest end of Yishan slope and the south of Tianhuan depression, which developed a set of alternate marine and continental coal-bearing formation during Carboniferous-Permian of Upper Paleozoic. According to the thermal simulation test results (Fig.4), coal rock has a certain amount of liquid hydrocarbon generation capacity when the Ro value is 1.0% to 1.3%, and the liquid hydrocarbon yield of different types of coal rock ranges from 50kg/t.TOC to 80 kg/t.TOC. Owing to coal rock's strong adsorption capacity, low hydrocarbon expulsion efficiency and high residual
hydrocarbon content, coal rock still contains a certain amount of soluble organic matter (chloroform bitumen "A" content is > 1000ppm) despite undergoing a high thermal evolution stage. These thermal cracked residual hydrocarbons accumulated in coal rock due to low hydrocarbon expulsion efficiency are cracked into gas under certain thermal evolution conditions. Moreover, there are also a small amount of residual soluble organic matter in dark mudstone and reservoir. The carbon isotope composition of the natural gas component generated by the cracking of soluble organic matter in the gas source rock and reservoir is lighter, and it is mixed with the high thermal evolution "dry gas", whereas mixing with cracking gas of soluble organic matter has no obvious effect on its carbon isotope composition because of the high content of methane; C_{2+} heavy hydrocarbon components are due to the very low content, only mixed with a small amount of cracking gas of soluble organic matter to make its carbon isotope composition significantly lighter, resulting in complete inversions ($\delta^{13}C_1 > \delta^{13}C_2 > \delta^{13}C_3$) or partial inversions ($\delta^{13}C_1 > \delta^{13}C_2 < \delta^{13}C_3$).

![Fig. 3 Carbon isotopic distribution model of the Paleozoic natural gas components of different genetic types in the Ordos Basin](image)

Residual cracking gas of soluble organic matter, mixed with coal-formed "dry gas", makes the carbon isotope composition of C_{2+} heavy hydrocarbon components significantly negative, which needs certain geological conditions. That is, the gas source rock and reservoir still contain a certain amount of soluble organic matter after they come to the maximum buried depth and experience the highest paleogeotemperature, then later with the tectonic uplift and the progressive decrease of geotemperature, which still has the temperature conditions for the cracking of soluble organic matter at the stage of cracking of liquid hydrocarbon. The burial thermal evolution history shows that the Yishan slope reached its maximum burial depth at the end of the Early Cretaceous (Ren et al., 2007; Ren et al., 2017). Due to the high thermal evolution degree in Longdong area, the high temperature cracking in the deep buried stage makes the content of heavy hydrocarbon components such as ethane in natural gas extremely low. In the early stage of the subsequent uplifting, although the paleogeotemperature gradually decreases, it is still in the liquid hydrocarbon cracking stage, and the small amount of residual soluble organic matter in the gas source rock and reservoir will continue to be cracked into a certain amount of heavy hydrocarbon components with a lighter carbon isotope composition. The amount of natural gas produced in the process is finite, but due to the extremely low C_{2+} component content in the original natural gas, the mixing of a small amount of natural gas rich in $^{12}C$ and C_{2+} components can just make the carbon isotope composition of C_{2+} heavy hydrocarbon components of "cracked dry gas" in the
original gas reservoir significantly lighter, while the carbon isotope composition of methane does not change significantly, which may cause complete inversions ($\delta^{13}C_1 > \delta^{13}C_2 > \delta^{13}C_3$) or partial inversions ($\delta^{13}C_1 > \delta^{13}C_2 < \delta^{13}C_3$).

**Fig. 4** Thermal simulation hydrocarbon yield curve of the Upper Paleozoic coal in the Ordos Basin

4. Conclusions

The Upper Paleozoic gases from the Longdong areas in the Ordos Basin are characterized by low contents of heavy hydrocarbon gases, the mean value of wetness as 2%, relatively heavy $\delta^{13}C_1$ value ($-28.3‰$), and large variations in $\delta^{13}C_2$. The distribution pattern of carbon isotopic sequence of natural gas is mainly characterized by the abnormal light $\delta^{13}C_2$ values and carbon isotopic reversal. According to the organic geochemical characteristics of natural gas and the development of gas-source rocks, the Upper Paleozoic natural gas in Longdong area is mainly derived from alternate marine and continental coal-bearing formation, which belongs to high mature and over-mature "coal-formed gas". The Ordovician strata are thin and the marine source rocks are underdeveloped in Longdong areas. In theory, only the mixture of high mature gas with extremely low contents of heavy hydrocarbon gases and a small amount of natural gas with higher contents and lighter carbon isotopic composition of heavy hydrocarbon gases can lead to the occurrence of abnormally light $\delta^{13}C_2$ and the carbon isotopic reversal between methane and ethane. The mixed gas is probably derived from the thermal cracking of a small amount of residual soluble organic matter in the gas-source rocks and reservoirs during the initial stage of the uplift movement at the end of Early Cretaceous.

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