Geochemical Characteristics of Natural Gas from Yangtake-Yudong2-Quele Structural Belt in Kuqa Depression, Tarim Basin, NW China

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ABSTRACT

This paper is to probe into the origin and maturity of Tertiary and Cretaceous natural gas samples from the Yangtake-Yudong2-Quele structural belt in Kuqa Depression of Tarim Basin, NW China, based on the analyses of the molecular and stable isotope compositions. The chemical composition data of 22 samples reveals that the gases mainly consist of hydrocarbon gas(exceed 95% often), lower heavy hydrocarbon gas contents(11.4% on average), and an average drying coefficient of about 88.39%, representing typical wet gas. Non-hydrocarbon gases in them, low in content, are mainly nitrogen and carbon dioxide, with nitrogen content much higher than carbon dioxide content. The stable carbon isotopes of 7 samples indicate alkane carbon isotope is heavy, especially of $\delta^{13}C_{2+}$（$\delta^{13}C_2$>-28‰）; with the increase of carbon number, the carbon isotope gets heavier, basically indicating the positive carbon isotope series of organic origin gas. The phenomenon of weak inversion of heavy hydrocarbon isotopes is common, probably caused by the mixing of gases of 'same source but different stages' or 'same stage but different sources'. By contrast methods and 3 classic graphic plates, the genetic type of the natural gas on the research belt is identified as humic pyrolysis gas, derived from thermal degradation of type Ⅲ kerogen. The vitrinite reflectance of the natural gas is between 1.01%-1.07%, calculated by the formula of $\delta^{13}C_1$-Ro and inferred by $\Delta(\delta^{13}C_1-\delta^{13}C_2)$ index. Compared with the published gas data from the Kelasu belt located in the northern depression, the natural gas of Yangtake are wetter (lower dry coefficient) and lighter (lighter carbon stable isotope), affected by the distance from the depression center and maturity.

KEYWORDS: Yangtake-Yudong2-Quele structural belt; Natural gas components; Natural gas stable carbon isotope; Genetic type; Maturity; Tarim Basin

INTRODUCTION

Kuqa Depression(KD), located at the northern margin of Tarim Basin, next to Tianshan fold belt on the north and North Tarim Uplift on the south, is the earliest first-order tectonic unit found in Tarim Basin, with the total natural gas reserves of $4422\times10^{8}m^{3}$ discovered by early 2013[1].

Yangtake-Yudong2-Quele structural belt (referred to as 'Yangtake' for short below) is located at the southwest margin of KD, situated at the west part of North Tarim Uplift, adjacent to northeast Yaha structural belt and southeast Yingmai 7 structural belt [2]. Well Yudong2 stands in the southwest of Yangtake belt and Quele-1 structure located at the west of the central section of Qiulitag belt, both having faulted anticlines (Figure 1).

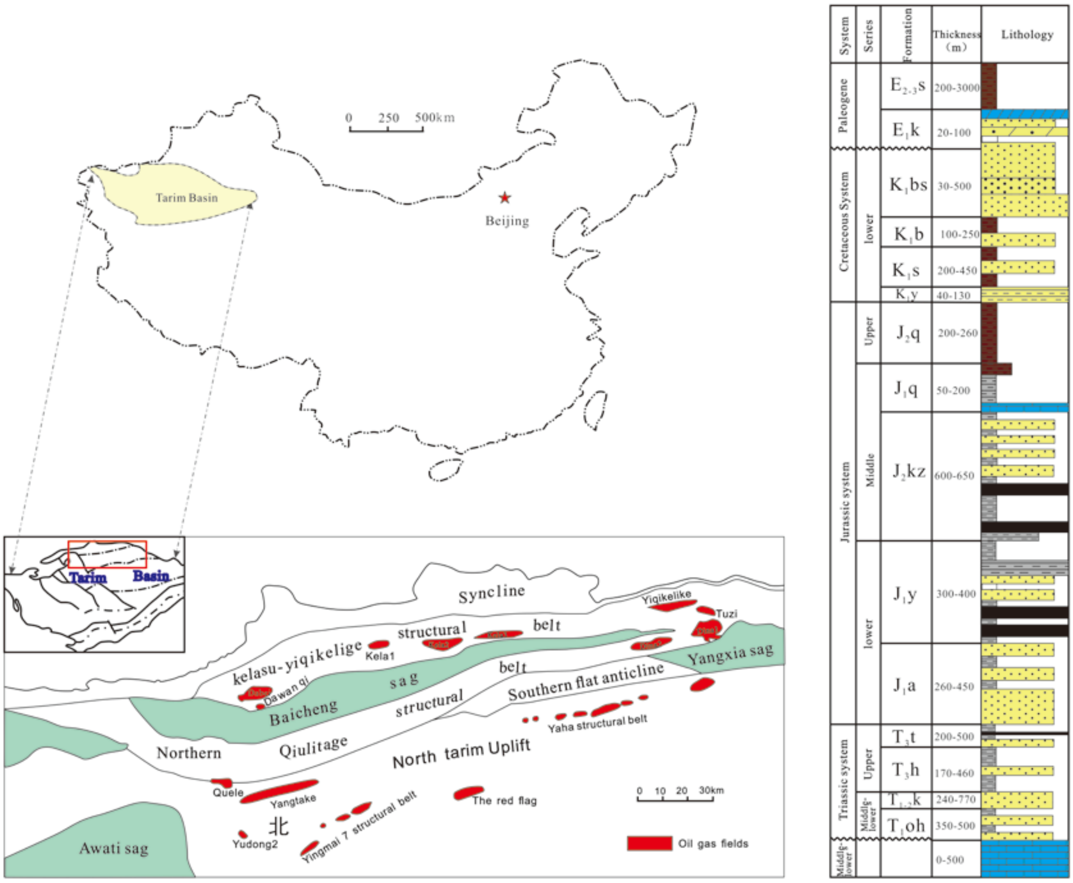


Figure 1: Distribution of structure units and stratigraphy in Kuqa Depression (modified after Zhou et al.[3])

Yangtake petroleum occurrences are parts of the frontal uplift belt of Kuqa petroleum system[4] which occurs in ring shape, developed periphery condensate reservoirs, with Jurassic-Triassic source rock and reservoir-caprock combination of lower Tertiary gypsum cover [5] and lower Tertiary, Cretaceous sandstones[6]. The traps bearing oil and gas are rich of fault blocks and fault nose structures[2] and the migration pathways consist of faults, unconformable surfaces and sandstone carrier beds[4].

Considerable and detailed literatures have revealed the geochemical characteristics of natural gas in Tarim Basin [3, 5, 7-11], however, there is no systematic study on gas geochemical traits in the Yangtake area. In order to better understand the properties of the gas reservoir, the geochemical characteristics of 22 natural gas samples taken from this belt were analyzed to figure out the genesis and maturity, coupling with previous published research results on dry gas from Kelasu structure, also within KD. The paper may help to understand the formation and distribution of gas reservoirs in the peripheral regions of KD, and provide geochemical basis for oil-gas exploration and prospective evaluation in this area.

METHODS AND RESULTS

A suit of 22 gas samples from 7 wells, 17 gases from Yangtake, 3 gases from Quele and 2 samples from Yudong, were collected. All samples were measured for natural gas composition. Among of seven samples, including 6 from Yangtake belt and 1 from Yudong belt, were obtained for carbon isotope test. All measurements were performed at the Key Laboratory of Exploration Technologies for Oil and Gas Resources, Ministry of Education, Yangtze University, from the PDB standard, on a Isoprime gas stable isotope mass spectrometer (GV company, UK) with a precision of ±0.2 ‰.

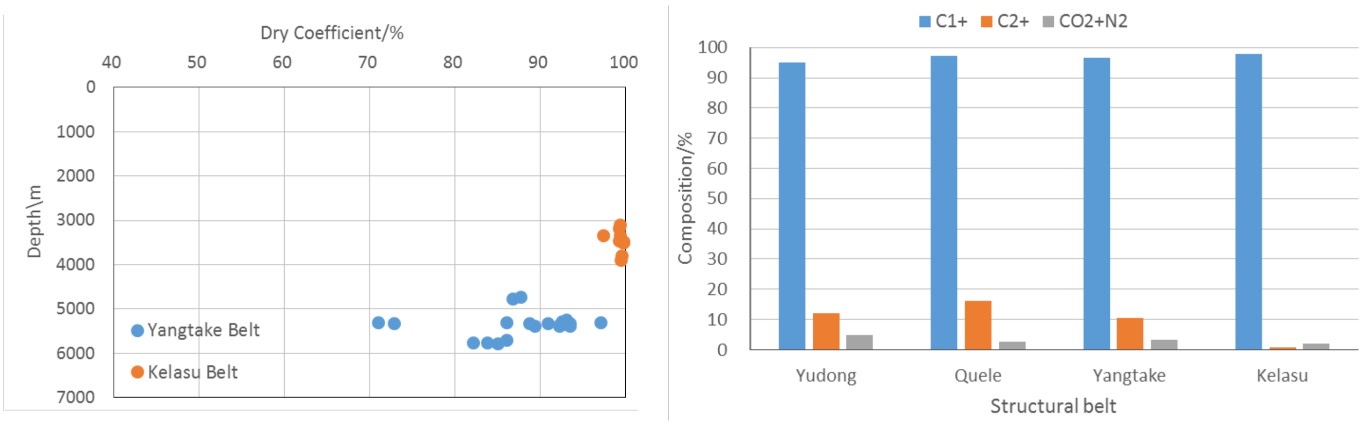
Natural gas compositions

Natural gas is composed of hydrocarbon gas and non-hydrocarbon gas[12]. The features of components are affected by gas genesis, maturity and differentiation or fractionation caused by migration or diffusion. The gas composition data acquired from tests and previous literature are presented in Table 1.

Results revealed gas components from Yangtake belt mainly consist of hydrocarbons(C_1-C_5) and non-hydrocarbon gas such as carbon dioxide (CO_2) and nitrogen (N_2), with the hydrocarbon gas taking the absolute dominance. The gas have contents of C_1-C_5 of more than 90% generally, and most samples higher than 96%. Gas samples with hydrocarbon contents between 90% and 95% account for 23%, and the gas samples from Well Yangta2 has the highest hydrocarbon content of 98.83% on average. Methane contents of the samples vary from 75% to 95%, and samples with methane content higher than 75% account for 91%. Few samples have methane contents lower than 70%, and mainly come from Well Yangta 5. Heavy hydrocarbon gas contents range between 2.92% and 27.99%, on average 11.35%, and samples with heavy hydrocarbon gas content of less than 12% account for 63.60%. Dry coefficients (ratio of C_1/(C_1-C_5)) vary in a wide range between 70.97% and 97.02%, on average 88.29%. All these indicate that the tested samples are quite different in composition and they are typical wet gas because their dry coefficients are lower than 95% (the boundary between dry gas and wet gas). The natural gas from Well Yangta5 has the lowest average dry coefficient (79.64%) and the Well Yangta2 has the highest (93.46%). The gas samples have very low non-hydrocarbon gas content, and trace amounts of other components. The non-hydrocarbon gas is mainly composed of CO_2 and N_2. Most of the samples have CO_2 content lower than 2%, and only 10% of the samples have CO_2 content of higher than 2%. The contents of N_2 vary between 0.09% and 7.86%, with the main frequency between 0% and 4%, which is apparently higher than CO_2.

Where possible, we compare composition data with published data from Kelasu belt [13], located in the northern KD (Table 1). The gas from Kelasu belt has higher methane content and dry coefficient, and much lower heavy hydrocarbon and non-hydrocarbon contents (Figure 2). In KD, the methane content of natural gas tends to decrease, and the C_2+ heavy hydrocarbon and non-hydrocarbon contents increase from the northern Kelasu to the southern Yangtake. Obviously, the differences in gas composition and variation tendency are controlled by the distance from the depression center and source rock maturity. Natural gas from Kelasu belt closer to the depression center is typical dry gas with higher maturity, methane content, and dry coefficient (＞0.95). In contrast, the natural gas from Yangtake belt farther away from the depression center is typical wet gas with relatively lower maturity and higher heavy hydrocarbon content.

Table 1 – **See the addendum**.



(a) (b)

Figure 2: (a)Dry coefficient with depth of Yangtake and Kelasu belts, (b)comparison of natural gas composition of different structure belts.

Natural gas carbon isotope

Through extensive studies, there is a consensus as to the carbon isotope of hydrocarbon, carbon isotope difference of methane and ethane ($\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ and alkane series can been viewed as important geochemical indexes for defining genesis, maturity of natural gas and gas source correlation. Carbon isotope of methane ($\delta^{13}C_1$) can be used to effectively determine the maturity of natural gas, while carbon isotope of ethane ($\delta^{13}C_2$) can indicate genetic type of organic matter in the gas source rock[12] . Δ ($\delta^{13}C_2$-$\delta^{13}C_1$) providing information on the relative change of carbon isotope, is another effective index of natural gas maturity[14, 15]. For a certain type of organic matter, it generally decreases with the increase of thermal evolution degree. When combining with $\delta^{13}C_1$ of methane, it can also be used to identify the genetic type of natural gas[15]. Alkane series can also be used to determine the genetic type of natural gas. The organic isotope values of organic originated alkane increase with carbon number, $\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3 < \delta^{13}C_4$; while the alkane of inorganic origin is opposite, $\delta^{13}C_1 > \delta^{13}C_2 > \delta^{13}C_3 > \delta^{13}C_4$. Irregular occurrence of $\delta^{13}C$ values of alkane is the so-called isotope reversal. Through systematic researches, Dai et al[12] summarized the five main causes for isotope reversal: ①mixing of organic alkane and inorganic alkane; ②mixing of coal-type alkane and oil-type alkane; ③mixing of residual natural gas generated at lower thermal maturity and natural gas generated at higher thermal maturity; ④biological degradation and ⑤increasing geo-temperature.

The gas carbon isotope data from Yangtake belt are summarized in Table 1. As comparison, the analysis results of carbon isotope of natural gas samples in Kelasu belt are also presented from the published literature. The histogram of carbon isotope distribution of the two structural belts (Figure 3) reveals that carbon isotopes from two belts are relatively high, especially $\delta^{13}C_{2+}$ values all spanning over -25‰.For Yangtake belt, the methane carbon isotope ($\delta^{13}C_1$) ranges between -42.4‰ and -36.2‰, on average -38.4‰; the ethane carbon isotope ($\delta^{13}C_2$) spans from -25.4‰ to -21.5‰ with a mean value of -23.4‰; the propane carbon isotope ($\delta^{13}C_3$) covers from -26.5‰ to -20.9‰ (average -24.1‰); the butane carbon isotope ($\delta^{13}C_4$) changes from -27.1‰ to -22.08‰ with an average of -24.3‰. For Kelasu belt, methane gas carbon isotope ($\delta^{13}C_1$) ranges between -31.5‰ and -26.91‰, on average -28.6‰; the ethane carbon isotope ($\delta^{13}C_2$) spans from -22.9‰ to -18.1‰ with a mean value of -25‰; the propane carbon isotope ($\delta^{13}C_3$) covers from -19.21‰ to -17.1‰ (average -20‰); the butane carbon isotope ($\delta^{13}C_4$) changes from -20.98‰ to -17.8‰ (average -19.9‰).

Table 1: Carbon isotope（‰）results of gas samples from Yangtake and Kelasu belts
See the addendum.

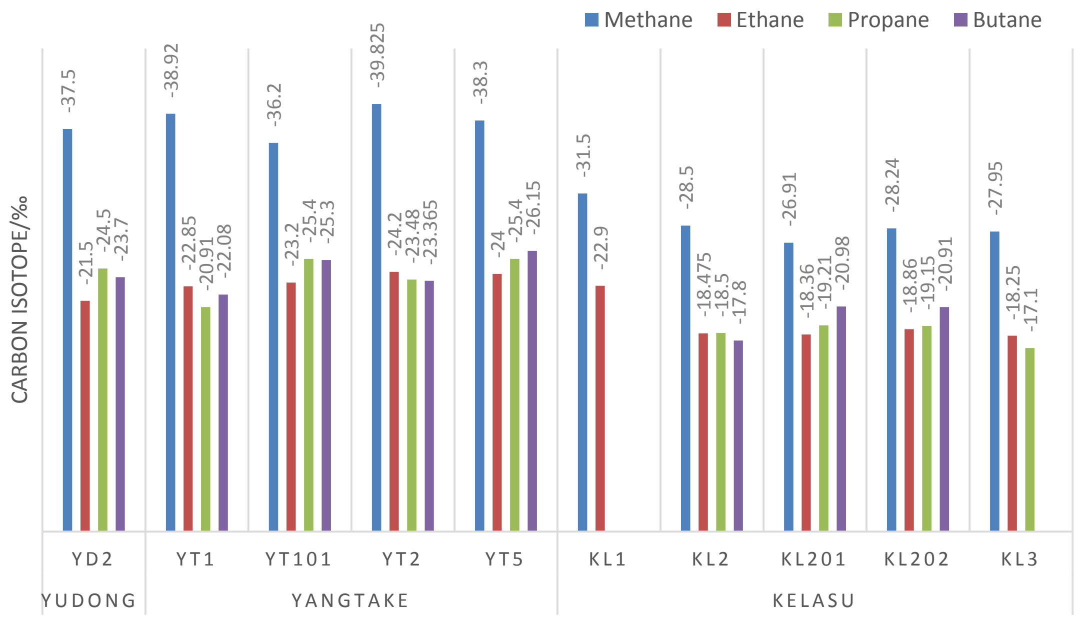


Figure 3: Histogram of carbon isotope distribution of natural gas samples from the study belt and Kelasu belt

Carbon isotope of natural gas generally becomes higher from Yangtake belt in the north to Kelasu belt in the south (Figure 3), especially for methane carbon isotope, increaseing from -38.4‰ of Yangtake to -28.6‰ of Kelasu. This variation reveals that methane carbon isotope is sensitive to thermal activity and has the most obvious isotope fractionation effect during thermal evolution. Carbon isotope fractionation also occurs in ethane, propane, and butane but with less in amplitude, only between 4‰ and 6‰. The fractionation of alkane carbon isotope becomes more severe with the increase of dry coefficient, methane content and decrease of heavy hydrocarbon (C_2^+) gas and non-hydrocarbon gas.

Carbon isotope of natural gas samples from the main exploration wells (Figure 4) show that the alkane series of the study belt are all series of heavy hydrocarbon's carbon isotope reversal, while the carbon isotope reversal of most gas samples from Kelasu belt feature $\delta^{13}C_2 > \delta^{13}C_3$ and a few $\delta^{13}C_3 > \delta^{13}C_4$. Slight carbon isotope reversal of heavy hydrocarbon gas is very common both in the study belt and Kelasu belt, which may be caused by mixing of natural gas generated either from a single set of source rocks at different evolution stages or from different sets of source rocks at the same stage.

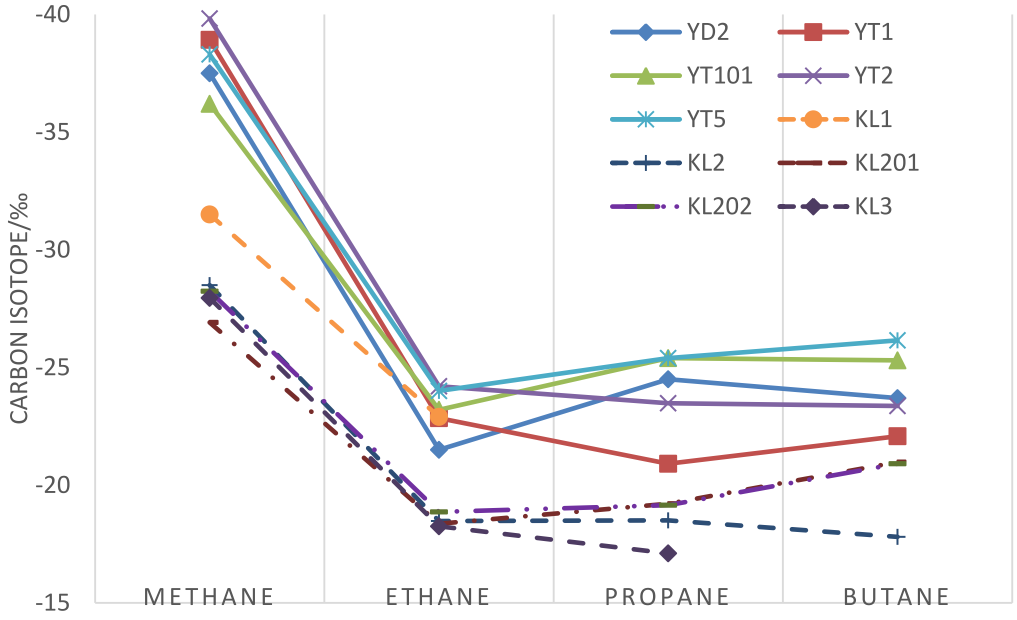


Figure 4: Carbon isotope of alkane series of natural gas samples from the Yangtake and Kelasu belts

Because the natural gas in KD accumulated in late period, the gas is mainly generated by Jurassic coal-bearing formation. The thick coal-bearing strata lead to difference in thermal maturity between source rocks at upper and lower part, and then carbon isotope reversal occurs when natural gases of different maturities mix. Condensate gas is the main type of natural gas developed in the frontal uplift of Yangtake belt and the natural gas and its associated condensate oil are inconsistent in origin, that is, they came from different source rocks. The formation of the associated condensate oil is related to the evaporative fractionation effect caused by injection of natural gas in late period.

Analysis results (Table 1) and distribution histogram (Figure 4) of natural gas carbon isotope also show that for Yangtake belt, the differences $\Delta(\delta^{13}C_1\text{-}\delta^{13}C_2)$ between carbon isotope of methane and ethane of gas samples are larger, between -16.95‰ and -13‰, with a mean value of -15.0‰; the carbon isotope differences between ethane, propane and butane, $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_3)$, are smaller; especially $\Delta(\delta^{13}C_3\text{-}\delta^{13}C_4)$, are usually calculated as positive values. Compared with Yangtake belt, $\Delta(\delta^{13}C_1\text{-}\delta^{13}C_2)$ of natural gas samples from Kelasu belt are relatively lower, ranging between -11.1‰ and -8.6‰, with a mean value of -9.38‰. The carbon isotope differences between ethane, propane and butane arealso usually positive. The differences between $\delta^{13}C_1$ and $\delta^{13}C_2$ indicate that during transition of Yangtake belt's wet gas to Kelasu belt's dry gas, $\delta^{13}C_1$ increase significantly with thermal maturity while the increase rate of $\delta^{13}C_2$ is smaller, and that's why $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ decreases with the increase of thermal maturity and dry coefficient.

In brief, in Yangtake belt, the carbon isotope of natural gas is generally heavy, especially for heavy hydrocarbon (C_2+). With the increase of carbon number, carbon isotope become heavier, which is an indication of organic genetic natural gas with positive carbon isotope series.

Controlled by distance from the center of depression and maturity of natural gas (reflected by dry coefficient and methane content), the carbon isotope becomes lighter from north to south, which is the same as natural gas composition. The closer to the depression center, the higher the maturity, the higher the dry coefficient and the higher the methane content of natural gas will be. At the same time, the fractionation effect of carbon isotope increasing with maturity becomes more obvious, especially, methane carbon isotope.

DISCUSSION

Origin of natural gas

Many kinds of methods can be used for genetic classification of natural gas. Based on extensive researches [12, 16], natural gas can be divided into 3 types: inorganic gas, organic gas and hybrid gas; organic gas was subdivided into coal type gas and oil type gas according to the types of parent material; and it also could be divided into biogenic gas, pyrogenic gas and cracked gas according to their thermal evolution degree. Pyrogenic gas and cracked gas can be subdivided into coal/oil derived pyrogenic gas and coal/oil derived cracked gas. The genetic type of natural gas can be identified by the composition of carbon isotopes of methane and its homologues[17]. Humic organic matter is poor in hydrogen and rich in aromatic condensation structure, rich in ^{13}C, and thus $\delta^{13}C$ is heaver. In contrast, sapropelic organic matter is rich in hydrogen and aliphatic chain structure, rich in ^{12}C, and $\delta^{13}C$ is lighter.

Identification of sapropelic gas and humic gas according to the kerogen type

Coal type gas refer to humic gas generated by thermal degradation of humic(Ⅲ) or near humic(Ⅱ$_2$) kerogen, not only to gas generated by coal[12, 18]. The source rocks of coal type gas include coal, carbonaceous mudstone and so on. Oil type gas refers to sapropelic gas generated by thermal degradation of type Ⅰ or type Ⅱ$_1$ kerogen [12, 18]. The types of natural gas origin are identified by contrast method and classic graphic plate in this essay.

Contrast method

In contrast method, the type of natural gas origin is identified by comparing measured carbon isotopes with standard carbon isotopes of oil type gas or coal type gas. A large number of gas source rock simulation experiments and research cases by Chinese researchers show that the composition of carbon isotopes of gas varies a lot with the types of organic matter in source rocks, which is mainly observed in hydrocarbons heavier than ethane [7]. So the values of carbon isotopes of ethane and propane can be used to differentiate natural gas into coal type gas and oil type gas, but the dividing lines are different between different researchers (Table 2). The value of methane carbon isotope mainly depends on thermal evolution degree and the type of gas source rock, it decreases with the increase of source rock quality, and increases with the rise of evolution degree. In 1987, Zhang, Gao and Jiang [19] reported that $\delta^{13}C_1 > -50‰$, $\delta^{13}C_2 < -29‰$ indicated oil type gas, and $\delta^{13}C_1 > -50‰$, $\delta^{13}C_2 < -29‰$ indicated coal derived gas. In 1991, Zhang [20] proposed that ethane carbon isotope was a good indicator of source rock type in the mature and high mature stage of organic matter, when condition evolution degree had less effect. He also thought $\delta^{13}C_2$= -28±1.5‰ could be taken as a basic line to distinguish sapropelic gas and humic gas. Dai [12] suggested to distinguish oil type gas and coal type gas with ethane and propane carbon isotopes, for oil type gas, the value of ethane carbon isotopes was lower than -28.8‰, and the value of the propane carbon isotopes was lower than -25.5‰; for coal type gas, the value of ethane carbon isotopes was higher than -25.1‰, and the value of the propane carbon isotopes was lower than -23.2‰. In 1996, Feng [17] classified gas with $\delta^{13}C_2$＞-29‰ as coal type gas, and gas with $\delta^{13}C_2$＜-29‰ as oil type gas. In 1997, Gang, Gao and Hao [21] confirmed by simulation experiments that carbon isotopes of kerogen had an strong inheritance, they could be inherited to ethane. He advancedthat $\delta^{13}C_2$＞-29‰ indicated humic gas, and $\delta^{13}C_2$＜-29‰ sapropelic gas. Liang, Zhang and Zhao [22] thought coal type gas had $\delta^{13}C_2$＞-28‰, $\delta^{13}C_3$＞-26‰, $\delta^{13}C_4$＞-25‰, oil type gas had $\delta^{13}C_2$＜-28‰, $\delta^{13}C_3$＜-26‰, $\delta^{13}C_4$＜-25‰。

Table 2: Identification standard of coal/oil type gas using carbon isotopes（‰）

	Index	Coal type gas	Oil type gas
Zhang [19]	$\delta^{13}C_1$	$\delta^{13}C_1$＞-50‰	$\delta^{13}C_1$＞-50‰
	$\delta^{13}C_2$	$\delta^{13}C_2$＜-29‰	$\delta^{13}C_2$＜-29‰
Zhang [20]	$\delta^{13}C_2$	$\delta^{13}C_2$＞-28+1.5‰	$\delta^{13}C_2$＜-28-1.5‰
Dai [12]	$\delta^{13}C_1$	-10＞$\delta^{13}C_1$＞-43	-30≥$\delta^{13}C_1$＞-55
		-10＞$\delta^{13}C_1$＞-43	
	$\delta^{13}C_2$	＞-25.1	＜-28.8
	$\delta^{13}C_3$	＜-23.2	＜-25.5
Feng [17]	$\delta^{13}C_2$	$\delta^{13}C_2$＞-29‰	$\delta^{13}C_2$＜-29‰
Gang [21]	$\delta^{13}C_2$	$\delta^{13}C_2$＞-29‰	$\delta^{13}C_2$＜-29‰
Liang [22]	$\delta^{13}C_2$	＞-28	＜-28
	$\delta^{13}C_3$	＞-26	＜-26
	$\delta^{13}C_4$	＞-25	＜-25

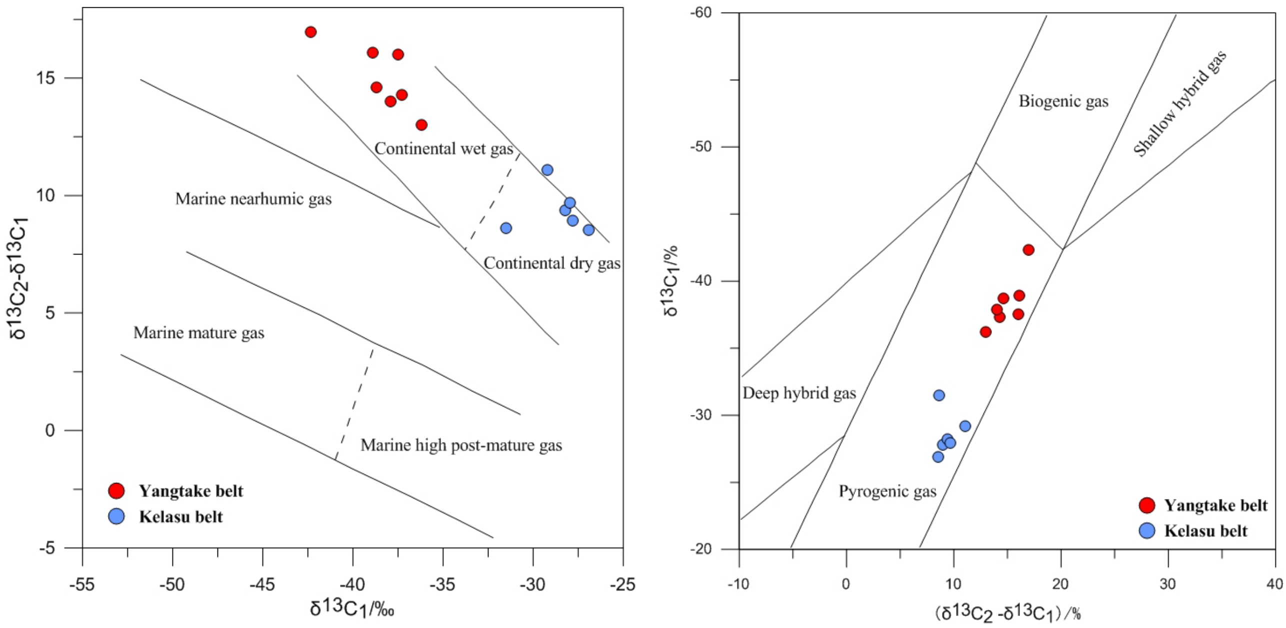
The ethane carbon isotope $\delta^{13}C_2$ in Yangtake belt is between -25.4‰ and -21.5‰, on average -23.4‰, according to the classification standards above, gas in Yangtake belt is classic humic (coal type) origin gas. Whereas from the composition of carbon isotopes in Kelasu belt, the ethane carbon isotope $\delta^{13}C_2$ is between -22.9‰ and -18.1‰, with an average of -19.22‰, which also indicates coal type humic gas. This is consistent with the lacustrine deposits in the Triassic-Jurassic strata of this depression.

Gas origin identification with $\Delta(\delta^{13}C_2-\delta^{13}C_1)-\delta^{13}C_1$ graphic plate

Zhao and Zhang [5] divided gas type of Tarim Basin into continental origin gas and marine origin gas by using $\Delta(\delta^{13}C_2-\delta^{13}C_1)$ and $\delta^{13}C_1$ according to the traits of components and carbon isotopes of natural gas. The continental origin gas, which corresponds to coal type gas, includes humic wet gas and humic dry gas. The marine origin gas, which corresponds to oil type gas, includes mature gas, high-post mature gas and partial humic mature gas. As illustrated in Figure

5-a, the carbon isotopes of 7 gas samples from Yangtake belt indicate they are continental wet gas, while 6 gas samples from Kelasu belt are continental dry gas, of humic coal derived gas origin.

Zhang [20] proposed the methane carbon isotope is the most important factor to define natural gas origin. Biogenic gas and bacterial decomposition gases generated by microbial activity have $\delta^{13}C_1$ < -55‰ generally, and deep source gas has $\delta^{13}C_1$ > -20‰, while pyrogenic gas has $\delta^{13}C_1$ between -55‰ and -20‰. Zhang [20] established an X type graphic plate for identifying natural gas origin according to the relationship of ($\delta^{13}C_2$-$\delta^{13}C_1$) and $\delta^{13}C_1$ (Figure 5-b). Using this graphic plate, the gases in the Yangake and Kelasu structural belts were identified as thermogenic gas too.



(a) (b)

Figure 5: Classification of natural gas origin on Yangtake belt according to classification plate of (a)Zhao and Zhang [5], Zhao [13] , (b) Zhang [20].

Gas origin identified by $\delta^{13}C_2$ *-*$\delta^{13}C_1$ *graphic plate*

Zhang [19] built the $\delta^{13}C_1$-$\delta^{13}C_2$ graphic plate based on methane and ethane carbon isotopes after analyzing more than 200 samples, which enables gas comparison and source rock tracking. According to this plate, oil type gas has $\delta^{13}C_1$ > -50‰ , $\delta^{13}C_2$ < -29‰ generally, coal type gas has $\delta^{13}C_1$ < -50‰ and $\delta^{13}C_2$ > -29‰. Biogenic gas has $\delta^{13}C_1$ < -55‰. Sapropelic biogenic gas has lighter $\delta^{13}C_2$, while humic biogenic gas has heavier $\delta^{13}C_2$, the dividing line of them is about -31‰ to -29‰. Coal bed methane has $\delta^{13}C_1$ between -71‰ and 32‰ and $\delta^{13}C_2$ > -29‰. According to this graphic plate, Figure 6 shows gases in Yangtake and Kelasu belts are humic thermogenic gas. But they have some differences in maturity, gas in Yangtake is high mature gas, while gas in Kelasu is post-mature gas.

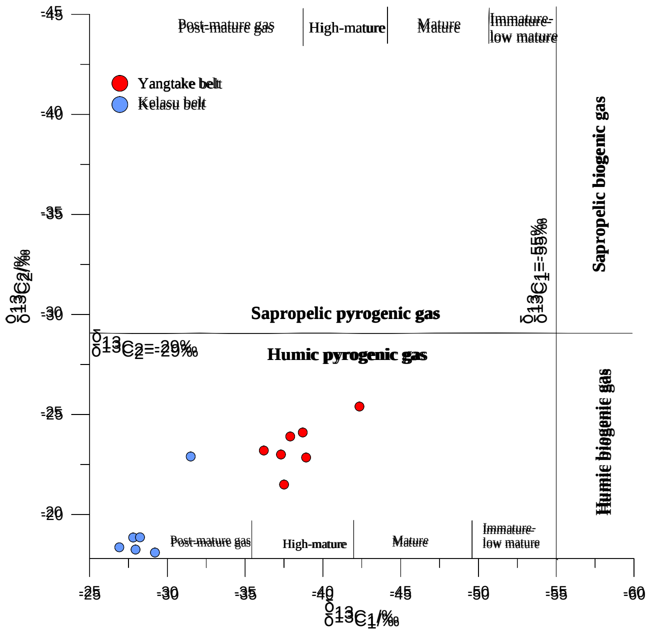


Figure 6: Identificationof origin of natural gas in Yangtake Belt (plate of Zhang [19])

Gas origin identification with $\delta^{13}C_1$-$\delta^{13}C_2$-$\delta^{13}C_3$ organic gas identification plate (V type plate)

Based on a lot of research on natural gas from China, Dai [12] proposed the $\delta^{13}C_1$-$\delta^{13}C_2$-$\delta^{13}C_3$ V type plate for natural gas origin identification. Adding the data of $\delta^{13}C_1$, $\delta^{13}C_2$ to the V type plate, Figure 7 shows the result points of the research belt fall mainly in the coal derived gas area, while $\delta^{13}C_1$ and $\delta^{13}C_3$ fall mainly in coal derived gas and hybrid gas area. Therefore, it is concluded that most gas samples in the research belt are coal derived gas, and a few coal derived gas and hybrid gas.

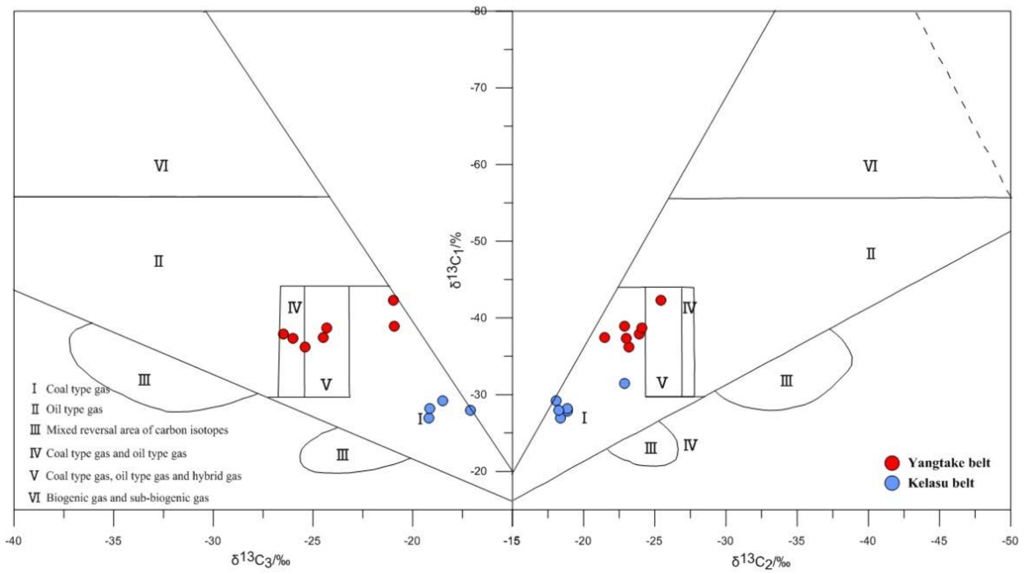


Figure 7: Identification of organic gas based on methane, ethane and propane

Taken together the results above by using graphic plates and contrast method, we can assume that the gas from Yangtake belt is coal type gas, and probably at the high mature stage, meanwhile, some mixed gas of ‘same type and different sources’ or ‘same source and different stages’ existing, with the kerogen type of Ⅲ.

Classification of coal type pyrogenic gas and coal type cracked gas

Pyrogenic gas refers to gas derived by organic thermal degradation in the mature and high mature stage (with Ro of 0.6%-2.0%). According to the kerogen type, pyrogenic gas can be divided into oil type pyrogenic gas and coal type pyrogenic gas. Cracked gas refers to gas derived by thermal cracking of residual kerogen, formed liquid hydrocarbon and a part of heavy hydrocarbon gas in the post-mature stage (Ro＞2.0%). According to the kerogen type, cracked gas can be divided into oil type cracked gas and coal type gas.

Coal type pyrogenic gas is derived by thermal catalysis of source rock, at Ro between 0.6% and 2.0%, corresponding to the stage of long flame coal to lean coal. Coal type pyrogenic gas has methane content of more than 80%, heavy hydrocarbon content of more than 5%, dry coefficient ($C_1/C_{1\text{-}5}$) of 0.70-0.95, C_2/C_3 of 0.8-3.0, iC_4/nC_4 of significantly less than 1, rich ^{13}C in methane, $\delta^{13}C_1$ between -45‰ and -30‰, and $\delta^{13}C_2$＞-28‰～-29‰. Coal type cracked gas is derived by thermal cracking of kerogen at Ro＞2.0%, corresponding to the stage over meagre coal, with methane content of greater than 95%, dry coefficient ($C_1/C_{1\text{-}5}$) of 0.95-1, C_2/C_3 of 1.5-7.0, rich ^{13}C in methane, and $\delta^{13}C_1$>-30‰ [7].

According to the standards above, the gas in Yangtake belt with an average methane content of 85%, heavy hydrocarbon content of 11%, dry coefficient ($C_1/C_{1\text{-}5}$) of 0.71-0.97, C_2/C_3 of 1-5, iC_4/nC_4 of 0.9, abundant ^{13}C in methane, $\delta^{13}C_1$ between -42‰ and -36‰, and the average $\delta^{13}C_2$ of -23‰, is coal type pyrogenic gas.

Maturity of natural gas

Maturity is an important geochemical index for natural gas reservoir research. Natural gas is formed throughout the process of organic matter evolution, but natural gases produced at different stages of evolution often have different geochemical properties. At the biochemical stage, the methane generated has very light $\delta^{13}C_1$ from -55‰ to -75‰[23] due to the selective intake of isotopes ^{12}C by methane genic bacteria. At the stage of pyrolysis, due to isotope fractionation, the $\delta^{13}C_1$ in the early formation of methane is lighter. With the increase of evolution degree of organic matter in source rock, $\delta^{13}C_1$ gradually becomes heavier and wider in range, ranging between -78‰ and -22‰. Therefore, $\delta^{13}C_1$ is a good indicator to identify the gas generation degree and the evolution of parent materials.

Identification of maturity according to δ13C1-Ro

Stahl, Boigk and Wollanke [24] have studied the natural gas in northwest Europe and North America, and they found that there is a logarithmic linear relationship between maturity(Ro) with the methane carbon isotope $\delta^{13}C_1$, moreover, the carbon isotope of coal derived gas in the same evolution degree is higher than that of oil type gas. But this is not suitable for China's natural gas. Since the 1980s, worldwide researchers like Schoell [25], Faber, Gerling and Dumke [26], Berner and Faber [27], Berner and Faber [28] and so on, have put forward the relationship between the $\delta^{13}C_1$ and Ro of coal type gas and oil type gas. Xu, Shen and Liu [29], Shen, Xu and Bin [30], Dai [12], Chen, Zhang and Xu [31], Li [9], Liu and Xu [32], Bao [7] and other researchers in China calculated the carbon isotopes and maturity formulas of methane by using the method of hydrocarbon generation kinetics and thermal simulation experiments(Table 3).

Since different formulas are applicable for specific areas, the relationship between methane carbon isotope composition and the corresponding vitrinite reflectance Ro (Table 5) obtained by Bao [7] thermal simulation experiments on mudstone and coal samples from KD was used to calculate natural gas maturity of the Yangtake research belt and the Kelasu structural belt in this study. The different source rocks in KD interbeded with each other and almost generated and expelled gas under the same conditions, both source rocks contributed to natural gas accumulation, it is hard to separate them strictly. Therefore, the average value of the two parameters is used as the index to measure the maturity of natural gas.

Table 3: Calculation formulas of $\delta^{13}C_1$ and maturity

See the addendum.

Table 5: Statistics of $\delta^{13}C_1$ and maturity of gas from Yangtake belt and Kelasu belt

Belt	Well	Horizon	Depth	$\delta^{13}C_1$	Vitrinite reflectance Ro,%		
			m	‰	Mudstone	Coal	Average
Yudong	Yudong 2	E-K	4728.81-4744.8	-37.5	1.09	1.04	1.07
Yangtake	Yangta 1	E+K	5234.37-5331.92	-38.92	1.00	0.93	0.97
	Yangta 101	E	5329-5333	-36.2	1.18	1.14	1.16
	Yangta 2	K	5387-5390	-37.3	1.11	1.05	1.08
	Yangta 2	K	5327.5-5401.75	-42.35	0.82	0.73	0.77
	Yangta 5	E	5310-5315	-37.9	1.07	1.01	1.04
	Yangta 5	E	5294.38-5321	-38.7	1.02	0.95	0.98
	Average				1.04	0.98	1.01
Kelasu	KL1	N	1469-1491	-31.5	1.57	1.61	1.59
	KL2	E	3499-3535	-29.2	1.81	1.91	1.86
	KL2	K	3803-3895	-27.8	1.96	2.12	2.04
	KL201	K	3630-4021	-26.91	2.07	2.26	2.17
	KL202	N	1472-1481	-28.24	1.91	2.05	1.98
	KL3	E	3104-3550	-27.95	1.95	2.09	2.02
	Average				1.88	2.01	1.94

The calculation results (Table 5) show that, the average value of the maturity of the oil type gas is 1.04%-1.09% in the Yangtake belt, the average value of the maturity of the coal derived gas is 0.98%-1.04%, and the average value of them is the maturity of the natural gas, i.e. Ro of

1.01%-1.07%. From the theory of oil and gas generation stage, it is found that the natural gas is in the mature stage. The mean value of the maturity of oil type gas is 1.88% in the Kelasu belt, and the average value of the maturity of coal type gas is 2.01%. The average value of them is the maturity of natural gas on the belt, Ro of 1.94%, indicating the high and post mature stage. Comparison of the maturity of the two belts shows maturity of natural gas in the Kuqa Depression gradually reduces from north to south.

Identification of maturity according to $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$

$\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ is an index for identifying the maturity of natural gas. Rooney [36] suggested that the carbon isotope difference between methane and ethane could be used to tell the maturity of natural gas. Studies by James [37] and Schoell [25] show that the carbon isotope difference between natural gas n-alkanes was not related to the source materials, but was affected by the maturity. $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ decreases with the increase of maturity, because the gas produced at the high mature stage (instant) has higher proportion of methane with heavier $\delta^{13}C_1$ value, and its accumulation causes heavier $\delta^{13}C_1$; while the $\delta^{13}C_2$ is mainly related to the parent materials type, with small increment of carbon isotope and absolute amount far less than methane, so the difference between the two tends to decrease and even reverses. Huang [35] pointed out that the difference of $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ gradually decreased with the increase of natural gas maturity, and was about 5‰-12‰ in the high mature stage (Ro=1.5% -2.4%), and decreased in the post-mature stage (Ro=2.4%-2.6%), even became negative (-2‰ to 5‰).

According to reversal variation of $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ and Ro proposed by Huang [35], the maturity stage of gas samples from different belts can be inferred. $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ of Yangtake gas samples are 13‰-16.95‰（average 15.0‰）, larger than 5‰-12‰ of high mature stage (Ro=1.5%-2.4%), so it is deduced that the gas from Yangtake belt is in the mature stage (Ro=1.0%-1.5%). The $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ of gas samples from Kelasu belt are lower smaller, 8.6‰-11.1‰, with an average of 9.38‰, which fall in the range (5‰-12‰) of post-mature stage (Ro=1.5%-2.4%). Therefore, the gas from Yangtake belt should be in the post-mature stage (Ro=1.5%-2.4%). There is a good negative correlation between the $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ and the calculated Ro scatter plot the gas samples from the research belt and Kelasu belt(Figure 8).

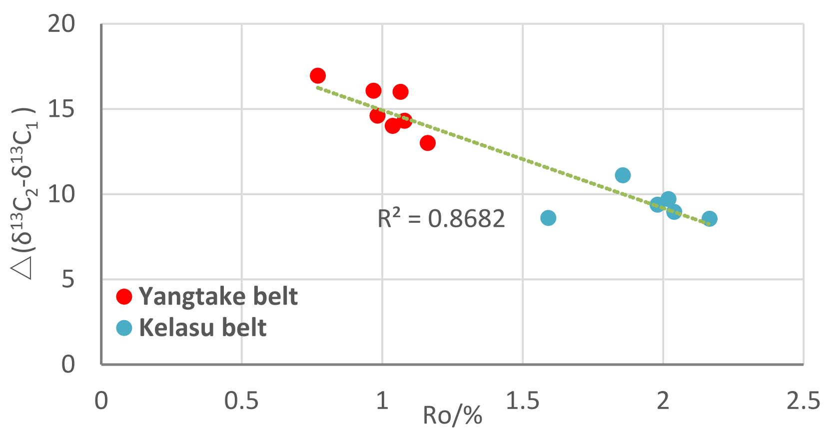


Figure 8: $\Delta(\delta^{13}C_2\text{-}\delta^{13}C_1)$ and Ro of Yangtake and Kelasu belts

CONCLUSIONS

(1) The gas samples from Yangtake-Yudong 2-Quele belt have definitely dominant hydrocarbon gas content, most of samples of over 95%, lower heavy hydrocarbon gas contents of 2.92-27.99%, average drying coefficient of 88.3%, representing typical wet gas. Nitrogen and carbon dioxide are two major non-hydrocarbon components in the natural gas, and the content of nitrogen is clearly higher than content of carbon dioxide. The weak reversal of heavy hydrocarbon isotopes are common, probably caused by the mixture of gases of ‘same source but different stages’ or ‘same stage but different sources’ of multiple gas source beds.

(2) The stable carbon isotopes of gas from the research belt show alkane carbon isotope is heavy ($\delta^{13}C_2 > -28‰$). By contrast method and three classic graphic plates ($\Delta(\delta^{13}C_2-{}^{3}C_1)-\delta^{13}C_1$, $\delta^{13}C_2-\delta^{13}C_1$, $\delta^{13}C_1-\delta^{13}C_2-\delta^{13}C_3$), it is confirmed the genetic type of natural gas on the research belt is mainly humic pyrolysis gas, derived from thermal degradation of type Ⅲ kerogen; and the the vitrinite reflectance of natural gas is between 1.01% and 1.07% in the mature stage, calculated by the formula of $\delta^{13}C_1$-Ro and inferred by $\Delta(\delta^{13}C_1-\delta^{13}C_2)$.

(3) Compared with gas of Kelasu structural belt, gas from Yangtake belt in the southern Kuqa Depression is more wet and lighter, characterized by ‘north dry and south wet’ in dry coefficient, and ‘north heavy and south light’ in carbon isotopes. Controlled by the distance from the center of the depression and the maturity of the natural gas, the natural gas has lower maturity and dry coefficient, and lower methane content. The wetter the component, the less the fractionation effect of methane with the maturity, and the lighter the carbon isotope is.

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