Research Article

Kaijun Tan, Juan Chen*, Jun Yao, Qingpeng Wu, and Jianglong Shi

Geochemical characteristics and genesis of natural gas in the Upper Triassic Xujiahe Formation in the Sichuan Basin

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Abstract: In recent years, the geochemical characteristics, genesis and sources of natural gas in the Upper Triassic Xujiahe Formation in the Sichuan Basin have received extensive attention, but their genesis and sources are still controversial. In this study, taking the natural gas from the Xujiahe Formation in the Sichuan Basin as an example, the source and genesis of the natural gas have been systematically analyzed. The results show that the natural gas of the Xujiahe Formation in the Sichuan Basin is dominated by methane, followed by a small amount of CO_2 and N_2; only the southern Sichuan area contains a small amount of H_2S, which comes from the supply of the underlying carbonate source rocks. Except for the western Sichuan Basin, the drying coefficient of the natural gas is generally less than 0.95 (wet gas). Furthermore, the composition of the natural gas is mainly controlled by the maturity of source rocks. The carbon isotope of ethane in natural gas ranges from −33.9 to −21.5‰, and the hydrogen isotope of methane ranges from −188‰ to −151‰. The carbon and hydrogen isotope values are higher in the western Sichuan Basin than in the central, northeastern and southern Sichuan Basin. The identification of the origin of natural gas and the comparison of gas sources show that the natural gas in the Xujiahe Formation is mainly coal-derived gas from its own coal-measure source rocks; the natural gas in the northern part of the southern Sichuan Basin is oil-derived gas originating from the Changxing Formation and the Silurian marine source rocks; however, the natural gas in the northeastern Sichuan Basin is a mixture of coal-derived and oil-derived gases. In addition, the carbon and hydrogen isotopes in some natural gas samples from the Xujiahe Formation have inversions of $\delta^{13}C_1 > \delta^{13}C_2$, $\delta^{13}C_2 > \delta^{13}C_3$, $\delta^{13}C_3 > \delta^{13}C_4$, and $\delta D_2 > \delta D_3$, and the magnitude of the inversions is small. It is considered to be caused by the mixing of gases from the same source, as well as the mixing of coal-derived and oil-derived gases.

Keywords: Sichuan Basin, Xujiahe Formation, geochemical characteristics, carbon isotope, hydrogen isotope, genesis of natural gas, source of H_2S

1 Introduction

The Upper Triassic Xujiahe Formation in the Sichuan Basin has a long history of natural gas exploration [1,2]. After more than half a century, 32 gas fields in the Zhongba, Bajiaochang, Chongxi, Guang'an and Hechuan areas have been discovered in the Sichuan Basin [3–5]. The Upper Triassic Xujiahe Formation has become one of the important natural gas producing layers in the Sichuan Basin [6–8]. In recent years, the geochemical characteristics, genesis and sources of natural gas in the Xujiahe Formation in the Sichuan Basin have received extensive attention, but their genesis and sources are still controversial [9–12]. Some researchers believe that the natural gas in the Xujiahe Formation mainly comes from coal-derived gas from its own coal-measure strata or oil-derived gas from underlying source rocks, but some researchers believe that it has multi-source mixing characteristics. Previous studies mostly used the comparisons of natural gas composition and carbon isotope characteristics, and these studies were usually limited to a certain well block [12–15]. Therefore, existing studies lack a comprehensive comparison of the genesis of natural gas in different regions of the whole basin, and thus have not formed an overall understanding of the genesis of natural gas in the Xujiahe Formation at the basin scale. In this study, natural gas samples from

* **Corresponding author: Juan Chen,** Research Institute of Petroleum Exploration and Development Northwest Branch, Lanzhou, 730020, China, e-mail: chenjuan1673@126.com
Kaijun Tan, Jun Yao, Qingpeng Wu, Jianglong Shi: Research Institute of Petroleum Exploration and Development Northwest Branch, Lanzhou, 730020, China

different areas in the Sichuan Basin were collected, and the analysis of natural gas composition, carbon and hydrogen isotopes was carried out. At the same time, the reported geochemical data on the Xujiahe Formation gas samples from different regions were collected, and a new overall understanding of the origin of the Xujiahe Formation gas at the basin level was developed. This study can provide a reference for future natural gas exploration in Xujiahe Formation.

2 Geological background

The Sichuan Basin is located in the northwestern margin of the Upper Yangtze Craton and is a rhombus-shaped hydrocarbon-bearing basin extending along the northeast strike. The structural units in the basin include the Western Sichuan Foreland Depression Area, the Central Sichuan Gentle Fold Area, the Northern Sichuan Thrust Fold Area, the Eastern Sichuan High-steep Structual Area, the Southern Sichuan Low-steep Fold Area and the Southwestern Sichuan Uplift Area (Figure 1). The Upper Triassic Xujiahe Formation in the Sichuan Basin is divided into the Xu 1 Member to the Xu 6 Member from bottom to top. It is a set of terrigenous clastic deposits dominated by inter-marine and continental facies [16–18]. The sedimentary paleogeomorphology of the Xujiahe Formation is generally higher in the east and lower in the west. In the process of deposition and tectonic movements, it has the characteristics of "early depositional overlay, and middle and late uplift and denudation" [19–21]. The sedimentary thickness in the western areas is large, which can reach more than 3,000 m; while the sedimentary thickness in the northern, eastern and western areas is relatively small, usually 200–300 m.

The source rocks of the Upper Triassic Xujiahe Formation in the Sichuan Basin are mainly shale and thin coal seams developed in the Xu 1, Xu 3 and Xu 5 Members. In addition, dark mudstone and thin coal seams are also developed in the middle and lower parts of the Xu 2 and Xu 4 Members, and the middle and upper parts of the Xu 6 Member. The Xujiahe Formation source rocks are thicker in the western Sichuan Basin (up to 1,000 m), and thinner in the northeastern and southern Sichuan Basin (generally between 50 and 100 m). Moreover, the Xujiahe Formation source rocks are rich in organic matter, and the organic carbon content is generally 0.5–9.7%, with an average value of 1.96%. The organic matter types of the Xujiahe Formation source rocks are mainly Types II_2 and III. The source rocks of the Xu 1, Xu 3 and Xu 5 Members are superimposed with the reservoirs of the Xu

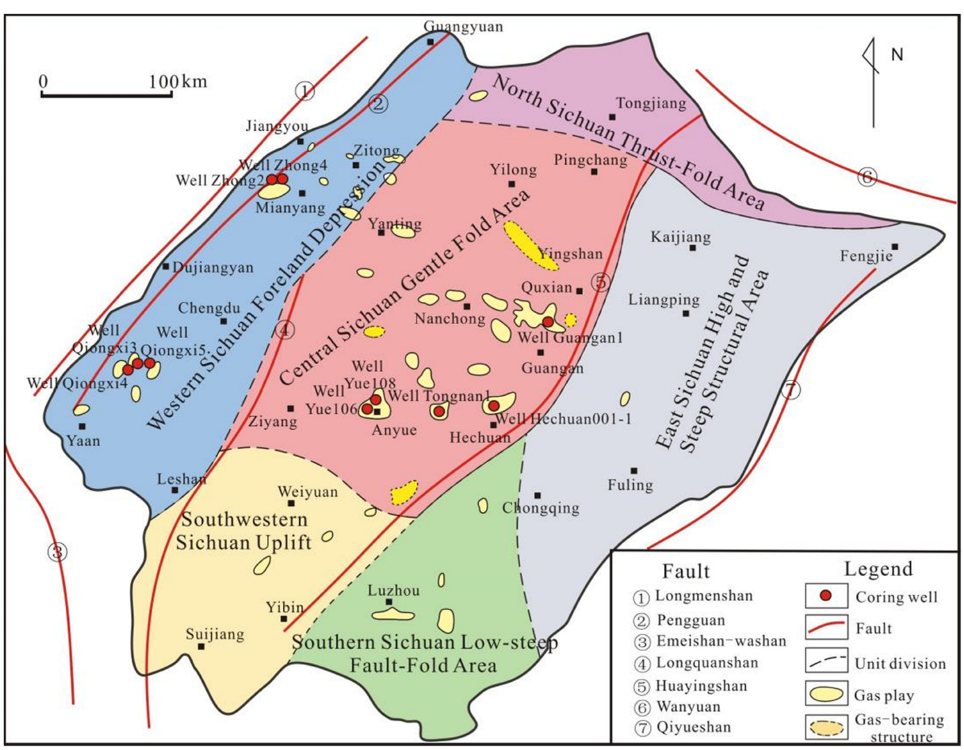

Figure 1: Structural unit division and gas play distribution in Sichuan Basin, China.

2, Xu 4 and Xu 6 Members, thus forming a good source-reservoir-caprock combination (Figure 2).

3 Experimental methods

Natural gas samples were collected from the Xujiahe Formation gas reservoirs in the central and western Sichuan Basin. Among them, the natural gas samples in the western Sichuan area were taken from the Zhongba 2 and Zhongba 4 wells in the Zhongba area; the Qiongxi 3, Qiongxi 4 and Qiongxi 5 wells in the Qiongxi area. The natural gas samples in the central Sichuan area were taken from the Guangan 1 well in the Guangan area; the Hechuan 001-1 well in the Hechuan area; the Anyu 106 and Anyue 108 wells in the Anyue area; and the Tongnan 1 well in the Tongnan area.

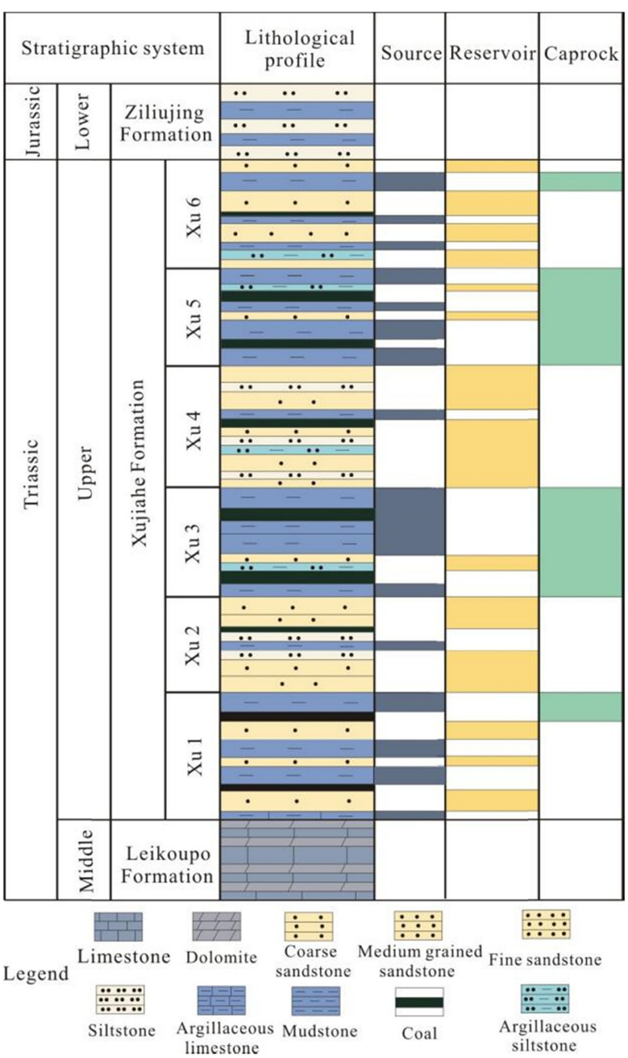

Figure 2: Comprehensive histogram of the Middle-Upper Triassic in the Sichuan Basin.

The sampling location was at the outlet of the well-head pressure gauge. In order to exclude the influence of external factors, the selected sample wells are normal production wells with long-term normal gas production and have not recently being added with chemical reagents.

Natural gas components were analyzed using an Agilent 7890 gas chromatograph. An gas chromatography-isotope mass spectrometry (GC-IRMS) was used for the carbon isotope analysis of natural gas. The device consists of a Thermo Delta V mass spectrometer connected to a Thermo Trace GC Ultra chromatograph with a PLOT Q column (27.5 m × 0.32 mm × 10 μm). In addition, the natural gas hydrogen isotope analysis was performed using a GC/TC/IRMS. The device consists of a MAT253 mass spectrometer connected to a Trace GC Ultra chromatograph equipped with a 1,450°C micro-pyrolysis furnace, and the chromatographic column is HP-PLOT Q (30 m × 0.32 mm × 20 μm). Each sample was tested at least 3 times, and the accuracy of the carbon isotope analysis was ±0.3‰ (VPDB standard), and the accuracy of the hydrogen isotope analysis was ±3‰ (VSMOW standard).

4 Results

4.1 Natural gas components

The natural gas in the Xujiahe Formation in the Sichuan Basin is dominated by hydrocarbon gas, and the total hydrocarbon volume is more than 95%. In the hydrocarbon components, CH_4 is absolutely dominant, and its volume is greater than 88%; while the non-hydrocarbon gas content is low, and the main components are mainly CO_2 and N_2. Only the natural gas of the Xujiahe Formation in the southern Sichuan Basin contains a small amount of H_2S, and the natural gas in the other regions usually does not contain H_2S. The drying coefficient of natural gas in the Xujiahe Formation is generally between 0.82 and 0.99, with an average value of 0.92 (Table 1). Among them, the drying coefficient of the western Sichuan Basin is generally high, up to 0.99, and the average value is 0.97, which is characterized by dry gas; while the drying coefficient of the central, northeastern and southern Sichuan Basin is lower than that of the western Sichuan Basin, and the average drying coefficients are 0.91, 0.90 and 0.89, respectively (wet gas).

Table 1: Component characteristics of natural gas in the Xujiahe Formation, Sichuan Basin

Areas	Volume fraction (%)				Drying coefficient	Data sources
	CH_4	C_2H_6	C_3H_8	H_2S		
Western Sichuan	81.14–98.31/94.15 (48)	0.51–7.83/2.66 (45)	0.01–5.18/0.65 (45)	0 (45)	0.89–0.99/0.97 (45)	[5,11]
Central Sichuan	80.16–97.82/89.66 (39)	0.54–11.37/5.91 (38)	0.01–6.25/1.34 (38)	0 (38)	0.82–0.99/0.91 (38)	[6,7]
Northeastern Sichuan	89.71–98.51/96.24 (9)	0.31–2.39/0.91 (9)	0.03–0.35/0.10 (9)	0 (9)	0.87–0.99/0.90 (9)	[19]
Southern Sichuan	81.87–93.25/88.05 (41)	0.58–10.57/6.13 (41)	0.04–4.81/2.21 (41)	0.03–0.54/0.21 (41)	0.85–0.99/0.89 (41)	[11]

Notes: min-max/average (number of samples).

On the whole, the variation trend of the drying coefficient of the Xujiahe Formation natural gas is basically consistent with the variation trend of the maturity of the Xujiahe Formation source rocks. The Xujiahe Formation in the western Sichuan area has a large burial depth and high maturity of source rocks, and its R_o is usually between 0.9 and 2.1%. For the central, northeastern and southern Sichuan areas, the burial depth of the Xujiahe Formation decreases, and the maturity of source rocks decreases as well. The R_o of the source rocks in the central Sichuan areas is between 0.8 and 1.6%, while the R_o of the source rocks in the southern Sichuan area is lower, mostly less than 1.2%.

4.2 Carbon isotope

From the samples collected in this study and the data of 48 samples collected in the references (Table 2), the distribution range of the carbon isotopes of methane in the natural gas is −42.5 to −30.0‰, with an average value of −36.0‰; the distribution of carbon isotopes of ethane ranges from −33.9 to −21.5‰, with an average value of −27.8‰. The carbon isotope characteristics of natural gas in different regions are different, and the western Sichuan region is higher than the central, northeastern and southern Sichuan regions. In the western Sichuan area, the distribution range of carbon isotopes of methane is −36.5 to −33.2‰, with an average value of −34.5‰, and the distribution range of carbon isotopes of ethane is −24.5 to −21.5‰, with an average value of −23.0‰. In the central Sichuan area, the distribution range of carbon isotopes of methane is −42.5 to −38.3‰, with an average value of −40.6‰, and the distribution range of carbon isotopes of ethane is −28.5 to −25.1‰, with an average value of −27.3‰. In the northeastern Sichuan area, the distribution range of carbon isotopes of methane is −36.5 to −30.0‰, with an average value of −31.6‰, and the distribution range of carbon isotopes of ethane is −33.9 to −30.3‰, with an average value of −32.2‰. In the southern Sichuan area, the distribution range of carbon isotopes of methane is −40.2 to −30.2‰, with an average value of −37.3‰, and the distribution of carbon isotopes of ethane is −33.8 to −26.5‰, with an average value of −28.8‰.

From the characteristics of the alkane gas carbon isotope series, most of the samples show a positive sequence of carbon isotopic composition ($\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3 < \delta^{13}C_4$). That is, the carbon isotope of alkane gas is more enriched in $\delta^{13}C$ with the increase of carbon

Table 2: Carbon isotopic characteristics of the natural gas in the Xujiahe Formation, Sichuan Basin

Areas	Gas-bearing structure	Well	Layer	δ13C‰							Data sources
				CH_4	C_2H_6	C_3H_8	C_4H_{10}	CO_2	N_2	H_2S	
Western Sichuan	Zhongba	Zhong2	T_3x_2	−35.4	−24.4	−23.1	−22.6	0.47	0.27		This study
		Zhong4	T_3x_2	−35.5	−24.5	−23	−22.5	0.46	0.31		This study
		Zhong19	T_3x_2	−35	−24	−22.5	−22.2	0.45	0.63		[18]
		Zhong34	T_3x_2	−35.4	−24.5	−22.8	−22.7	0.13	1.2		[18]
		Zhong36	T_3x_2	−35.4	−24.4	−22.9	−22.6	0.52	0.21		[18]
		Zhong44	T_3x_2	−35	−24	−22.7	−22.6	0.47	0.91		[18]
		Zhong63	T_3x_2	−35.5	−24.4	−23	−22.5	0.46	0.28		[18]
	Qiongxi	Qiongxi3	T_3x_2	−33.7	−22.4	−22.1	−21.9	1.55	0.23		This study
		Qiongxi4	T_3x_2	−33.5	−22.5	−22.3	−21.8	1.47	0.24		This study
		Qiongxi5	T_3x_2	−36.5	−24.2	−21.8	−21.1	0.07	0.53		This study
		Qiongxi6	T_3x_2	−34.5	−22.1	−22		0.92	0.21		[19]
		Qiongxi13	T_3x_2	−33.2	−21.5	−21.7		1.47	0.25		[19]
		Qiongxi16	T_3x_2	−34.1	−22	−21.7		1.39	0.20		[19]
	Pingluoba	Pingluo3	T_3x	−33.3	−21.7	−21.3	−20.3	0.76	0.54		[20]
		Pingluo6	T_3x	−33.5	−21.7	−22.6	−22.1	0.77	0.37		[20]
		Pingluo8	T_3x	−33.6	−21.6	−21.6	−20	0.73	0.48		[20]
		Pingluo10	T_3x	−33.7	−21.7	−22.7	−22.6	0.81	0.39		[20]
Central Sichuan	Bajiaochang	Jiao47	T_3x_6	−39.5	−25.1	−21.7	−24.1	0.19	0.19		[9]
		Jiao48	T_3x_6	−40.3	−26.5	−24.2	−22.7	—	0.67		[19]
		Jiao31	T_3x_4	−40.1	−27.4	−24.6	−24.6	0.54	–		[19]
		Jiao49	T_3x_2	−37	−27.3	−24.2	−22.9	—	0.11		[19]
	Chongxi-Lianchi	Xi20	T_3x_4	−42.5	−28	−25.2	−24.1	0	0.87		[17]
		Xi72	T_3x_4	−41.7	−28.3	−26	−24.9	—	—		[21]
		Xi73X	T_3x_4	−43.7	−28.4	−26.2	−25.5	0.28	0.80		[17]
		Lianshen1	T_3x_4	−40.5	−27.4	−24.5	−23.2	—	0.42		[17]
		Xi35-1	T_3x_2	−41.7	−28.3	−26	−24.9	0.25	1.37		[17]
	Guangan	Guangan1	T_3x_6	−39.4	−27.1	−25.4	23.8	0.36	0.77		This study
		Guangan2	T_3x_6	−39.7	−27.4	−25.7	−24.7	0	4.76		[17]
		Guangan51	T_3x_6	−39.5	−26.5	−25.5		0	0.67		[21]
		Guangan126	T_3x_6	−39	−26.3	−24.8	−22.2	0.37	0.49		[17]
	Hechuan	Hechuan001-1	T_3x_2	−39.6	−27.1	−23.9	−24.3	0.16	0.44		This study
		Hechuan106	T_3x_2	−39.8	−27	−24.1		0.21	0.39		[19]
		Hechuan108	T_3x_2	−41.4	−28.3	−25	−27.2	0.26	0.54		[19]
		Hechuan109	T_3x_2	−38.3	−26.2	−23.6		0.15	0.31		[19]
	Anyue	Yue101	T_3x_2	−41.3	−26.8	−23.7	−25.2	0.35	0.71		[19]
		Yue101-11	T_3x_2	−41.1	−26.3	−23	−25.1	0.30	0.43		[19]
		Yue105	T_3x_2	−41.6	−28.5	−25.4	−26.2	0.29	0.59		[19]
		Yue106	T_3x_2	−41.6	−27.5	−24.9	−25.8	0.20	1.28		This study
		Yue108	T_3x_2	−41.5	−27.1	−23.8	−25.4	0.28	1.37		This study
	Tongnan	Tongnan1	T_3x_2	−41.5	−27.6	−24.3	−26.9	0.11	1.00		This study
		Tongnan104	T_3x_2	−41	−27.4	−24	−26.7	0.76	0.43		[19]
		Tongnan105	T_3x_2	−40.4	−27.4	−24	−25.9	0.27	0.37		[19]
		Tongnan001-2	T_3x_2	−40.7	−27.5	−24.5	−26.1	0.30	0.39		[19]
Northeastern Sichuan	Yuanba	Yuanba1	T_3x_2	−31.8	−30.8			0.58	0		[22]
		Yuanlu8	T_3x_2	−30.4	−33.5	−33.5		62.83	15.33		[22]
		Yuanlu9	T_3x_2	−30	−33	−33.6		1.29	8.13		[22]
		Yuanlu10	T_3x_2	−31.5	−32.3	−32.7		2.56	0.05		[22]
	Tongnanba	Heba104	T_3x_4	−30.5	−32.2	−29.5		2.74	0.45		[16]
		Renhe1	T_3x_4	−30.6	−31.3			3.07	0.12		[16]
		Ma101	T_3x_4	−31.5	−33.9			0.24	0.36		[16]
	Wolonghe	Woqian1	T_3x	−36.5	−30.3	−25.3		1.01	0.44		[9]
Southern Sichuan	Guanyinchang	Yin10	T_3x_6	−38.5	−26.5	−23.3		0.63	0.31	0.18	[23]
		Yin17	T_3x_6	−40.2	−27.2	−23.7		—	0.64	0.13	[23]
		Yin27	T_3x_4	−38.8	−26.9	−23.5		0.03	0.35	0.21	[23]

(*Continued*)

Table 2: *Continued*

Areas	Gas-bearing structure	Well	Layer	δ13C‰							Data sources
				CH_4	C_2H_6	C_3H_8	C_4H_{10}	CO_2	N_2	H_2S	
	Hebaochang	Bao27	T_3x_2	−39.9	−28.3			012	1.31	0.36	[21]
	Danfengchang	Dan2	T_3x_2	−37.2	−28.6	−27.6		0.17	0.46	0.54	[21]
	Naxi	Naqian1	T_3x_6	−36.6	−30	−25.2		0.75	1.18	0.05	[21]
	Hejiang	He8	T_3x_6	−30.2	−33.8			0.24	0.41	0.03	[9]

number. This is consistent with the typical carbon isotopic composition characteristics of the prototype without modification and its origin. In some natural gas samples, the carbon isotopes of $\delta^{13}C_1 > \delta^{13}C_2$, $\delta^{13}C_2 > \delta^{13}C_3$, and $\delta^{13}C_3 > \delta^{13}C_4$ are inverted, and the inversion amplitude is small (<1‰). Moreover, the inversion amplitude of individual samples is relatively large, such as the inversion of $\delta^{13}C_3 > \delta^{13}C_4$ at angle 47, and the inversion amplitude reaches 2.35‰ (Figure 3). Among all the 21 samples with carbon isotope inversion, the number of samples with inversion in the western, central, northeastern and southern Sichuan areas were 5, 11, 4 and 1, respectively. The number of

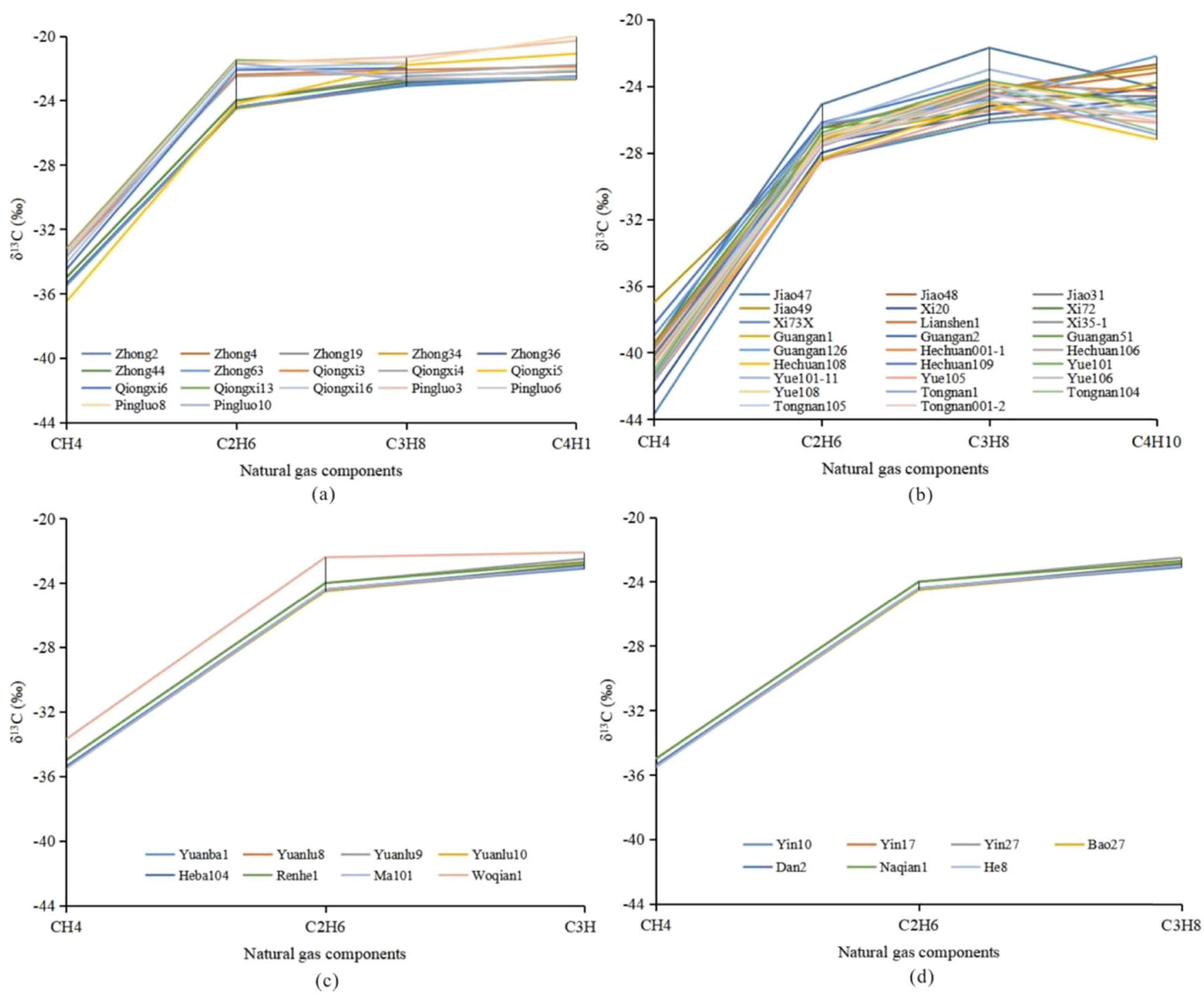

Figure 3: Distribution characteristics of carbon isotope series of natural gas in the Xujiahe Formation in Sichuan Basin. Notes: (a) Western Sichuan area; (b) Central Sichuan area; (c) Northeastern Sichuan area; (d) Southern Sichuan area.

samples with $\delta^{13}C_1 > \delta^{13}C_2$, $\delta^{13}C_2 > \delta^{13}C_3$ and $\delta^{13}C_3 > \delta^{13}C_4$ inversions were 3, 5 and 13, respectively. 13C-depleted is caused by the biochemical activity of ancient microorganisms when methane was consumed.

4.3 Hydrogen isotope

From the data of 10 samples collected in this study and 40 samples in the references (Table 3), the distribution of hydrogen isotopes of natural gas in the Xujiahe Formation in the Sichuan Basin ranges from −188 to −151‰, with an average value of −166‰. The hydrogen isotope characteristics of natural gas in different regions are different, and the hydrogen isotope value in the western Sichuan region is higher than that in the central, northeastern and southern Sichuan regions. Among them, in the western Sichuan area, the distribution range of the hydrogen isotope of methane is −171 to −157‰, with an average value of −165‰; in the central Sichuan area, the distribution range of the hydrogen isotope of methane is −188 to −156‰, with an average value of −177‰; in the northeastern Sichuan area, the distribution range of the methane hydrogen isotopes is −166 to −155‰, with an average value of −159‰; in the southern Sichuan area, the distribution range of the methane hydrogen isotopes is −183 to −145‰, with an average value of −165‰.

From the characteristics of the hydrogen isotope series of alkane gas, most samples show a positive sequence arrangement of hydrogen isotope composition ($\delta D_1 < \delta D_2 < \delta D_3$). The $\delta D_2 > \delta D_3$ inversions of natural gas in five samples in the western Sichuan area and two samples in the central Sichuan area occurred, and the inversion amplitudes were small; while the hydrogen isotopes in the northeastern and southern Sichuan areas did not show inversion (Figure 4). Among all the samples, only the carbon and hydrogen isotopes of the Well Qiongxi 13 were reversed at the same time.

5 Discussion

5.1 Origin of natural gas

Alkane carbon isotope is a common indicator for identifying the origin of natural gas. Therefore, the coupled analysis of various alkane carbon isotopes can help people obtain more accurate and comprehensive information on the origin of natural gas. First, we put all the sample data of the Xujiahe Formation into the $\delta^{13}C_1$–$\delta^{13}C_2$–$\delta^{13}C_3$ identification template (V-type identification template [24]). It can be seen from Figure 5 that the data in the western and central Sichuan Basin all fall into the Type I coal-derived gas area; the Yuanba and Tongnanba data in the northeastern Sichuan area mostly fall into the Type III carbon isotope inversion area, while the Wolonghe data fall in the interval of Type II oil-type gas; the data in the southern Sichuan Basin are distributed in these four types. On the whole, the natural gas data are mainly distributed in the coal-based gas regions, and a small part is distributed in the oil-derived gas regions. In addition, a small amount is distributed in the coal-derived gas and/or oil-derived gas area and the carbon isotope inversion area. This fully shows that the natural gas in this area is mainly coal-derived gas, and there is also a certain amount of oil-derived gas, rather than the typical coal-derived gas considered in previous studies.

The carbon isotopes of alkanes are controlled by the thermal evolution degrees of source rocks and the type of parent material. Among them, methane is greatly affected by the thermal evolution degree, while carbon isotopes of heavy hydrocarbon gases such as ethane are less affected by the thermal evolution degree. Carbon isotopes have strong inheritance of the original parent material, and are effective indicators for identifying coal-derived and oil-derived gases [25]. Wang's research on the geochemical characteristics of the Sinian-Jurassic natural gas in the Sichuan Basin believes that the value of $\delta^{13}C_2$ of coal-derived gas is greater than −29‰ [26]. Chen et al. conducted a comparative study on coal-derived and oil-derived gases in major petroliferous basins in China, and found that the value of $\delta^{13}C_2$ for coal-derived gas is greater than −28‰, while the value of $\delta^{13}C_2$ for oil-derived gas is less than −28‰ [27]. Dai et al.'s comprehensive research on the characteristics of natural gas in China found that the value of $\delta^{13}C_2$ for coal-derived gas is greater than −27.5‰, and the value of $\delta^{13}C_2$ for oil-derived gas is less than −29‰ [25]. Based on the discriminant indicators proposed by the previous and the geological background of the Sichuan Basin, this study uses $\delta^{13}C_2 = -29$‰ as the boundary between coal-derived and oil-derived gases in the Xujiahe Formation of the Sichuan Basin. That is, when the value of $\delta^{13}C_2$ is greater than −29‰, it is coal-derived gas, and when the value of $\delta^{13}C_2$ is less than −29‰, it is oil-derived gas. From the test data, the distribution range of $\delta^{13}C_2$ value in the western and central Sichuan Basin is −21.5 to −28.5‰, which are all greater than −29‰; while the distribution range of the $\delta^{13}C_2$ value in the northeastern and southern

Table 3: Hydrogen isotopic characteristics of natural gas in the Xujiahe Formation, Sichuan Basin

Areas	Gas-bearing structure	Well	Layer	δD‰			Data sources
				CH_4	C_2H_6	C_3H_8	
Western Sichuan	Zhongba	Zhong2	T_3x_2	−171	−145	−138	This study
		Zhong4	T_3x_2	−171	−144	−136	This study
		Zhong19	T_3x_2	−170	−144	−135	[18]
		Zhong34	T_3x_2	−170	−143	−135	[18]
		Zhong36	T_3x_2	−171	−143	−136	[18]
		Zhong44	T_3x_2	−171	−145	−137	[18]
		Zhong63	T_3x_2	−170	−145	−136	[18]
	Qiongxi	Qiongxi3	T_3x_2	−159	−134	−136	This study
		Qiongxi4	T_3x_2	−158	−133	−135	This study
		Qiongxi5	T_3x_2	−157	−135	−138	This study
		Qiongxi6	T_3x_2	−158	−132	−118	[19]
		Qiongxi13	T_3x_2	−158	−134	−137	[19]
		Qiongxi16	T_3x_2	−159	−134	−139	[19]
Central Sichuan	Bajiaochang	Jiao48	T_3x_6	−185	−153	−142	[19]
		Jiao31	T_3x_4	−182	−144	−138	[19]
		Jiao49	T_3x_2	−172	−144	−139	[19]
	Chongxi-Lianchi	Xi20	T_3x_4	−183	−140	−144	[17]
		Xi72	T_3x_4	−177	−137	−122	[21]
		Xi73X	T_3x_4	−186	−141	−147	[17]
		Lianshen1	T_3x_4	−173	−149	−129	[17]
		Xi35-1	T_3x_2	−156	−127	−107	[17]
	Guangan	Guangan1	T_3x_6	−180	−155	−131	This study
		Guangan2	T_3x_6	−184	−158	−134	[17]
		Guangan51	T_3x_6	−168	−127	−112	[21]
		Guangan126	T_3x_6	−171	−155	−140	[17]
	Hechuan	Hechuan001-1	T_3x_2	−175	−136	−126	This study
		Hechuan106	T_3x_2	−172	−129	−119	[19]
		Hechuan108	T_3x_2	−183	−135	−118	[19]
		Hechuan109	T_3x_2	−163	−136	−126	[19]
	Anyue	Yue101	T_3x_2	−188	−132	−125	[19]
		Yue101-11	T_3x_2	−178	−129	−117	[19]
		Yue105	T_3x_2	−183	−129	−119	[19]
		Yue106	T_3x_2	−181	−128	−116	This study
		Yue108	T_3x_2	−184	−131	−121	This study
	Tongnan	Tongnan1	T_3x_2	−176	−125	−117	This study
		Tongnan104	T_3x_2	−179	−128	−119	[19]
		Tongnan105	T_3x_2	−173	−128	−118	[19]
		Tongnan001-2	T_3x_2	−176	−123	−116	[19]
Northeastern Sichuan	Yuanba	Yuanlu8	T_3x_2	−160			[22]
		Yuanlu9	T_3x_2	−155			[22]
		Yuanlu10	T_3x_2	−157			[22]
	Nantongba	Renhe1	T_3x_4	−157			[16]
		Ma101	T_3x_4	−166			[16]
Southern Sichuan	Guanyinchang	Yin10	T_3x_6	−175	−129	−115	[23]
		Yin17	T_3x_6	−183	−134	−119	[23]
		Yin27	T_3x_4	−173	−131	−118	[23]
	Hebaochang	Baoqian4	T_3x_4	−145	−108	−93	[10]
		Baoqian1	T_3x_2	−172	−128	−107	[10]
		Bao27	T_3x_2	−156	−114	−105	[21]
	Danfengchang	Dan2	T_3x_2	−151	−136	−122	[21]

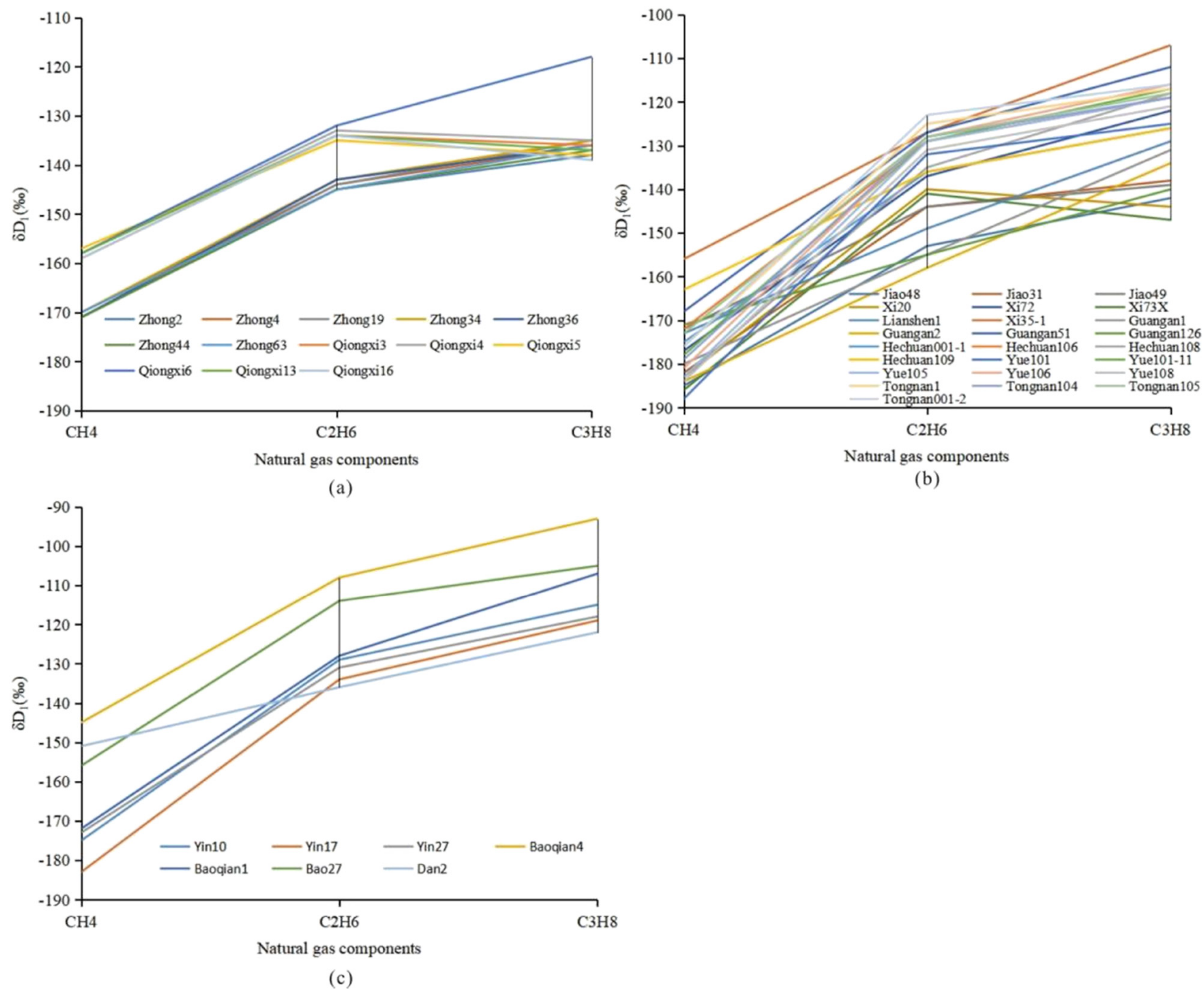

Figure 4: Distribution characteristics of hydrogen isotope series in natural gas of Xujiahe Formation in Sichuan Basin. Notes: (a) Western Sichuan area; (b) Central Sichuan area; (c) Southern Sichuan area.

Sichuan Basin is −30.0 to −40.2‰, which are all less than −29‰.

Whether the natural gas of the Xujiahe Formation in the northeastern and southern Sichuan Basin is oil-derived gas or a mixture of coal-derived and oil-derived gases has always been a hot topic of discussion. The study found that most of the data from the Xu 2 Member gas reservoir in the Yuanba Gas Field in the northeastern Sichuan Basin fall in the carbon isotope inversion area shown in Figure 5. The distribution range of the $\delta^{13}C_2$ value is −30.8 to −33.5‰, and the $\delta^{13}C_2$ value is less than −29‰. The natural gas generated from the Permian Wujiaping/Longtan Formation source rocks under the Xujiahe Formation has a heavier ethane carbon isotopic composition. For example, the $\delta^{13}C_2$ value of the natural gas in the Longgang Gas Field generated by the Permian Wujiaping/Longtan Formation source rocks ranges from −22.0 to −27.0‰ [9]. Therefore, it is impossible for their mixing to form natural gas with such a light carbon isotope of ethane. The deep Qiongzhusi Formation develops typical marine sapropelic source rocks, which are currently in the over-mature stage. Previous studies have shown that the Qiongzhusi Formation source rocks mainly generate crude oil cracked gas, and its associated pitch carbon isotope is distributed in the range of −33.1 to −35.4‰, and the ethane isotope of natural gas is generally lighter than −33‰. Therefore, the crude oil cracked gas from the Qiongzhusi Formation was mixed with the coal-derived gas from the Xujiahe Formation, and the mixed gas with the carbon isotope of ethane less than −29‰ was formed.

Since the Qiongzhusi Formation is deeply buried and is currently in the over-mature stage, it has the geological conditions for the formation of crude oil cracked gas.

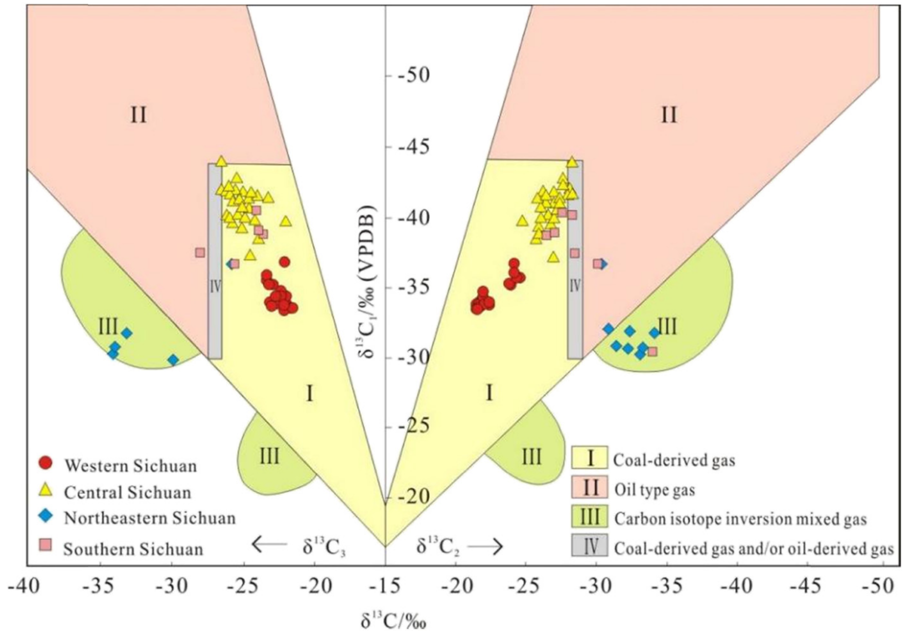

Figure 5: Distribution characteristics of $\delta^{13}C_1$–$\delta^{13}C_2$–$\delta^{13}C_3$ in the natural gas of the Xujiahe Formation, Sichuan Basin (refer to Dai et al. [24] for the bottom map).

Solid bitumen is an additional product of crude oil cracking in the Qiongzhusi Formation. The crude oil was cracked deep and migrated to the Xujiahe Formation along faults. It mixed with the Xujiahe Formation coal-derived gases, rather than being transported to the Xujiahe Formation for further cracking. Therefore, the carbon isotope of solid pitch of the Qiongzhusi Formation is lighter than that of ethane in natural gas. Overall, the natural gas in the gas reservoir of the Xu2 Member of the Xujiahe Formation in the Yuanba Gas Field mainly comes from the crude oil cracking gas generated from the source rocks of Xujiahe Formation and the marine source rocks of the underlying Cambrian Qiongzhusi Formation. It is a mixture of coal-derived and oil-derived gases.

According to the carbon isotopic characteristics of methane and ethane in natural gas in the Qiongzhusi and Xujiahe Formations, the mixing ratio of different types of gases can be calculated. The results show that the mixing ratio of the crude oil cracked gas in the Qiongzhusi Formation and the coal-derived gas in the Xujiahe Formation is 4:6.

The data of the Xu 6 Member gas reservoir in the Hejiang Gas Field in the southern Sichuan Basin fall in the carbon isotope inversion area in Figure 4. The $\delta^{13}C_2$ value is –33.8‰, and the $\delta^{13}C_2$ value is less than –29‰, which is close to the $\delta^{13}C_2$ value (–34.7 to –33.8‰) of the underlying Permian Maokou and Carboniferous Huanglong Formations [9]. The Xujiahe Formation faults in the Hejiang Gas Field are relatively developed, and most of them terminate upwards in the Upper Triassic Xujiahe Formation, and end downwards in the developed layers of carbonate gas reservoirs such as the Middle and Lower Triassic and Permian strata. Local faults break down to the Silurian, and these faults provide conditions for deep oil-type gas to enter the Xujiahe Formation gas reservoirs. Previous studies have shown that the natural gas in the Huanglong Formation and other gas reservoirs under the Xujiahe Formation in the Hejiang Gas Field comes from the contribution of the Silurian source rocks [28–33].

From the data of the Xu 4 Member gas reservoir in the Naxi Gas Field in the southern Sichuan Basin, the carbon isotopes of alkane gas are arranged in a positive sequence. The distribution range of its $\delta^{13}C_2$ value is –30.0 to –30.7‰, and the $\delta^{13}C_2$ value is less than –29‰, indicating that it is oil-derived gas. The carbon isotopes of alkane gas in the underlying Triassic Jialingjiang Formation gas reservoir are also arranged in positive order. The distribution range of $\delta^{13}C_2$ value is –32.1 to –33.2‰ [9], which is close to the $\delta^{13}C_2$ value of the Xujiahe Formation gas reservoirs. However, the carbon isotope of the alkane gas in the Lower Permian gas reservoir is reversed, showing a "V"-shaped arrangement (Figure 6). Its $\delta^{13}C_2$ value distribution range is –35.1 to –35.4‰ [9]. Therefore, the Xujiahe Formation gas reservoir in the Naxi Gas Field is of the same origin as the Jialingjiang

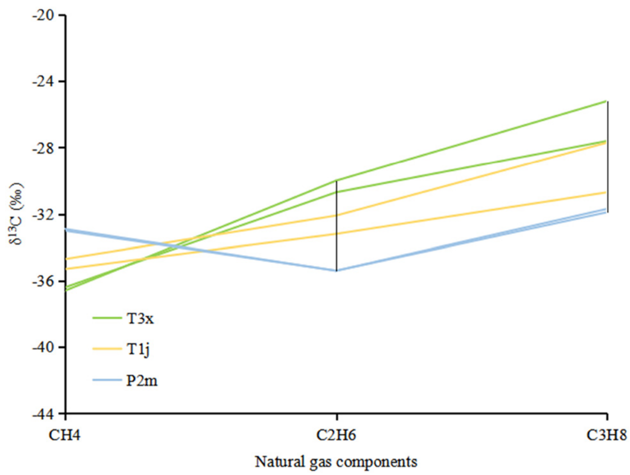

Figure 6: Distribution characteristics of carbon isotope series of natural gas in different layers of Naxi Gas Field in southern Sichuan Basin.

Formation gas reservoirs. According to previous studies, the natural gas in the Jialingjiang Formation gas reservoir may come from the argillaceous limestone source rocks of the underlying Changxing Formation, rather than the coal measure source rocks of the Longtan Formation [30].

Using ethane carbon isotopes to identify the origin and source of natural gas has certain limitations, especially for gas reservoirs formed by mixing coal-derived and oil-derived gases [34–37]. Heavy hydrocarbon gases such as methane and ethane may have different origins [38–40]. Therefore, there are some drawbacks in using only the carbon isotope characteristics of ethane to determine the origin of methane-based natural gas [6]. The hydrogen isotope of alkane gas is mainly controlled by the depositional environment and maturity of source rocks, and can be used as an important indicator to identify the depositional environment of source rocks, they provide an important supplement for the identification of the origin of natural gas [6,41–44]. Liao et al. used statistical methods to systematically study the hydrogen isotope characteristics of natural gas in the Sichuan Basin, and believed that the δD_1 value of the natural gas of marine and continental origins in the Sichuan Basin uses –150‰ as the dividing point. That is, the δD_1 value of marine gas is generally higher than –150‰, and the δD_1 value of continental gas is generally lower than –150‰ [31]. Yu et al. believed that the distribution range of δD_1 value of natural gas in marine strata in the Sichuan Basin is –129 to –120‰, while that of natural gas in the Xujiahe Formation is –180 to –160‰ [10]. According to Liu's research, the δD_1 value of marine natural gas in the Tarim Basin is basically higher than –160‰ [32]. In this study, δD_1 = –155‰ was adopted as the boundary between marine and continental origins in the Sichuan Basin. That is, when the value of δD_1 is greater than –155‰, it is of a marine origin, and when the value of δD_1 is less than –155‰, it is of a terrestrial origin. Figure 7 shows that the distribution range of δD_1 values in the western Sichuan Basin is –157 to –171‰, while that in the central Sichuan Basin is –188‰ to –156‰. The δD_1 values in these two areas are both less than –155‰, which indicate they are continental natural gases. The distribution range of δD_1 value in northeastern Sichuan Basin is –166 to –155‰, and that in the southern Sichuan Basin is –183 to –145‰. The values of some samples are greater than –155‰, indicating that the natural gas of the Xujiahe Formation in some areas has contributions from marine source rocks.

From the perspective of ethane carbon isotope, the ethane carbon isotope of the natural gas in the Xujiahe Formation in the central and western Sichuan Basin is generally heavier than –29‰, indicating that it is mainly from the continental humic source rocks of the Xujiahe Formation itself; however, the ethane carbon isotope of the natural gas in the Xujiahe Formation in the northeastern Sichuan Basin is generally lighter than –29‰, indicating that it has a contribution from marine sapropelic source rocks of the deep Qiongzhusi Formation. From the perspective of hydrogen isotope, the hydrogen isotope of the natural gas in the Xujiahe Formation in the central and western Sichuan Basin is less than –155‰, which is of continental origin. The hydrogen isotope values of some samples in the Xujiahe Formation natural gas in the northeastern Sichuan Basin are greater than –155‰, indicating that they have the supply of marine source rocks. Therefore, it is consistent with the conclusion of ethane carbon isotope analysis.

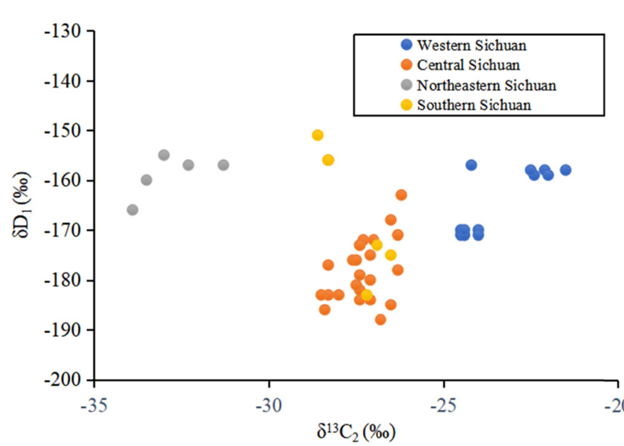

Figure 7: Correlation between δD_1–$\delta^{13}C_2$ of natural gas in the Xujiahe Formation, Sichuan Basin.

5.2 Origin of H_2S

The natural gas in the Xujiahe Formation in most areas of the Sichuan Basin does not contain H_2S, and only the Xujiahe Formation in the southern Sichuan area contains H_2S (Table 1). Generally, the presence or absence of H_2S content in natural gas is obviously controlled by the lithology of the reservoirs [9,45–47]. Natural gas in clastic rocks usually has low or no H_2S content, while natural gas in carbonate rocks generally contains H_2S, and sometimes at high levels. The reason is that, first, the sandstone has a large surface area and has a desulfurization effect, and H_2S cannot exist; and the sandstone is generally deposited in an oxidizing environment and contains more oxidants (Fe_2O_3, etc.), so even if H_2S accumulates in the sandstone, it is easily oxidized to form pyrite. Second, in carbonate rock formations, the TSR reaction is prone to occur to form H_2S [9], which leads to preservation in carbonate rock reservoirs lacking Fe_2O_3. The natural gas in the Xujiahe Formation in the western and central Sichuan Basin does not contain H_2S, indicating that the natural gas comes from its own coal-measure source rocks, without the supply of underlying carbonate source rocks. However, some natural gas in the Xujiahe Formation in the southern Sichuan Basin contains a small amount of H_2S, indicating that this part of natural gas comes from the supply of the underlying marine source rocks. There are three main sources of H_2S in natural gas [48–50]: (1) directly from the thermal cracking of kerogen or sulfur-containing compounds in crude oil; (2) from the bacterial reduction of sulfate; (3) from the thermochemical reduction of sulfate. In this study, marine carbonate formations are developed in southern Sichuan Basin, and the H_2S in natural gas is mainly derived from the TSR reaction of marine carbonate formations.

5.3 Reasons for partial inversion of carbon isotopes

There are five reasons for the inversion of carbon isotopes in alkane gas [6]: (1) the mixing of organic and inorganic alkane gases; (2) the mixing of coal-derived and oil-type gases; (3) the mixing of different source gases of the same type or different phases of the same source; (4) one or more kind of components of alkane gas are oxidized by bacteria; (5) thermochemical reduction of sulfate (TSR).

Some natural gas in the Xujiahe Formation in the central Sichuan area has partial inversed of carbon isotope. The structure of this area is stable, the faults are not developed, and the natural gas of the Xujiahe Formation in this area is mainly of crustal origin [6], so the possibility of the mixing organic and inorganic alkane gas is excluded. In addition, the natural gas in the Xujiahe Formation in this area is self-generated and self-storage coal-derived gas, and no oil-derived gas is mixed, so the influence of oil-derived gas mixing can be excluded. Moreover, the Xujiahe Formation is buried at a depth of more than 2,000 m, the temperature is relatively high, and there is no sign of oxidation and biodegradation. The carbon isotope of methane in the natural gas of bacteria decomposed gas is generally lighter than −55‰, indicating that there is no mixing of bacteria decomposed gas. At the same time, there is no sign of biodegradation according to the condensate and bitumen biometrics. In addition, the Xujiahe Formation in this area is composed of sandstone, dark mudstone and coal seams, and there are no sulfate sediments, so the effect of TSR on the carbon isotope of alkane gas can be excluded. Then, the reason for the carbon isotope inversion for some natural gas reservoirs in the Xujiahe Formation in this area is the mixing of the same type of gas from different sources or the same source gas from different periods. Previous studies have shown that there are early and late fluid inclusions in the Xujiahe Formation tight sandstone reservoirs in the central Sichuan Basin [30], which proves that the mixing of gas from the same source is the reason for the inversion of natural gas carbon isotopes in this area.

The natural gas of the Xujiahe Formation in the Yuanba and Tongnanba areas in the northeastern Sichuan region have obvious partial inversion or even continuous inversion of carbon isotopes in alkanes. This is because the natural gas in this area mainly comes from the crude oil cracking gas generated from the source rocks of the Xujiahe Formation and the marine source rocks of the underlying Cambrian Qiongzhusi Formation. Therefore, the inversion of carbon isotopes in natural gas in this area is caused by the mixing of coal-derived and oil-derived gases.

6 Conclusions

(1) The natural gas of the Xujiahe Formation in the Sichuan Basin is dominated by methane, followed by a small amount of CO_2 and N_2; only the southern Sichuan area contains a small amount of H_2S, which comes from the supply of the underlying carbonate source rocks. Except for the western Sichuan Basin,

the drying coefficient of natural gas is generally less than 0.95, and it shows the characteristics of wet gas. Furthermore, the composition of natural gas is mainly controlled by the maturity of source rocks.

(2) The carbon isotope of ethane in natural gas ranges from −33.9 to −21.5‰, and the hydrogen isotope of methane ranges from −188 to −151‰. The carbon and hydrogen isotope values are higher in western Sichuan Basin than in central, northeastern and southern Sichuan Basin.

(3) The identification of the origin of natural gas and the comparison of gas sources show that the natural gas in the Xujiahe Formation in the western Sichuan Basin is mainly coal-derived gas from its own coal-measure source rocks; the natural gas in the northern part of the southern Sichuan Basin is oil-derived gas originating from the Changxing Formation and the Silurian marine source rocks; however, the natural gas in the northeastern Sichuan Basin is a mixture of coal-derived and oil-derived gases.

(4) The carbon and hydrogen isotopes in some natural gas samples from the Xujiahe Formation in the Sichuan Basin have inversions of $\delta^{13}C_1 > \delta^{13}C_2$, $\delta^{13}C_2 > \delta^{13}C_3$, $\delta^{13}C_3 > \delta^{13}C_4$, and $\delta D_2 > \delta D_3$, and the magnitude of the inversions is small. It is considered to be caused by the mixing of gas from the same source, as well as the mixing of coal-derived and oil-derived gases.

Conflict of interest: Authors state no conflict of interest.

Data availability statement: Data are available by contacting the corresponding author.

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