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Geochemical Characteristics and Genetic Types of Natural Gas in the Xinchang Gas Field, Sichuan Basin, SW China

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Abstract: The molecular compositions and stable carbon and hydrogen isotopic compositions of natural gas from the Xinchang gas field in the Sichuan Basin were investigated to determine the genetic types. The natural gas is mainly composed of methane (88.99%–98.01%), and the dryness coefficient varies between 0.908 and 0.997. The gas generally displays positive alkane carbon and hydrogen isotopic series. The geochemical characteristics and gas-source correlation indicate that the gases stored in the 5th member of the Upper Triassic Xujiahe Formation are coal-type gases which are derived from source rocks in the stratum itself. The gases reservoired in the 4th member of the Xujiahe Formation and Jurassic strata in the Xinchang gas field are also coal-type gases that are derived from source rocks in the 3rd and 4th members of the Xujiahe Formation. The gases reservoired in the 2nd member of the Upper Triassic Xujiahe Formation are mainly coal-type gases with small amounts of oil-type gas that is derived from source rocks in the stratum itself. This is accompanied by a small amount of contribution brought by source rocks in the Upper Triassic Ma'antang and Xiaotangzi formations. The gases reservoired in the 4th member of the Middle Triassic Leikoupo Formation are oil-type gases and are believed to be derived from the secondary cracking of oil which is most likely to be generated from the Upper Permian source rocks.

Key words: natural gas, geochemical characteristics, genetic types, Xinchang gas field, western Sichuan Basin

1 Introduction

Natural gas is generally dominated by gaseous alkanes with relatively simple chemical compositions compared to crude oil. The geochemical characteristics of natural gas, such as chemical composition, carbon and hydrogen isotopic composition, as well as noble gas isotopic composition are fundamental for the study of the origin, migration, accumulation and alteration of natural gas, and have played an important role in revealing the organic type, thermal evolution degree and sedimentary environment of natural gas source rocks (Stahl, 1977; Schoell, 1980; Chung et al., 1988; Prinzhofer and Huc, 1995; Rooney et al., 1995; Whiticar, 1999; Tang et al., 2000; Zhang Xiaobao et al., 2003; Dai et al., 2004; Zhu Guangyou et al., 2007; Liu Quanyou et al., 2011; Ni et al., 2011; Liu et al., 2014a; Meng et al., 2015; Tao Cheng et al., 2015; Fu Xiaofei et al., 2016; Liu et al., 2016; Ma Yongsheng and Zhao Peirong, 2016).

The Sichuan Basin is one of the important petroliferous basins onshore in China, and the natural gas exploration in the basin has achieved significant success in recent years. The geochemical characteristics, origin and source of the gases have been widely studied. Several large gas fields such as the Xinchang, Guang'an and Hechuan have been explored in the terrigenous strata in central and western Sichuan Basin. It was identified that the natural gas reservoired in the Upper Triassic and Middle-Upper

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Jurassic layers was coal-type gas which was mainly derived from the coal-measure source rocks in the Upper Triassic Xujiahe Formation (Qin Shengfei et al., 2007a, 2007b; Dai et al., 2009, 2012; Wu Xiaoqi et al., 2011; Bian Congsheng et al., 2015; Yu Yu et al., 2016). The exploration in the marine strata in the Sichuan Basin has also made a significant breakthrough. On one hand, the exploration in the Upper Permian Changxing and Lower Triassic Feixianguan formations has discovered several large gas fields such as Puguang, Longgang and Yuanba in northeastern Sichuan Basin. The gases in these gas fields were commonly considered as oil-cracking gases that were derived from the secondary cracking of oil generated by source rocks in the Upper Permian Longtan Formation. They had generally experienced alteration by thermalchemical sulfate reduction (TSR) (Hao et al., 2008, 2015; Liu et al., 2013, 2014b).Whereas on the other hand, the exploration in the Upper Sinian Dengying and Lower Cambrian Longwangmiao formations has revealed that the Anyue giant gas field has reserves of over 400×10^9 m^3 in central Sichuan Basin. In addition, the gases were considered as oil-cracking gases derived from the Sinian and Cambrian source rocks (Wei Guoqi et al., 2015). Moreover, the shale gas exploration in the Lower Silurian Longmaxi Formation in southeastern Sichuan Basin has attracted wide attention (Dai et al., 2014). Therefore, the Sichuan Basin has become one of the focused areas in the exploration and geochemical study of natural gas in China.

The Xinchang gas field locates in the central section of western Sichuan Basin, which has been previously documented about its reservoir property and its main controlling factors (Lin Xiaobing et al., 2014; Wang Zhengrong et al., 2015; Wu Xiaoqi et al., 2016). Few studies have been conducted on the geochemical characteristics of natural gas with no consensus on the gas origin and source. Natural gas reservoired in the 4th member of the Middle Triassic Leikopou Formation was considered as either oil-type gas derived from source rocks in the Leikoupo Formation (Xie Gangping, 2015) or coal-type gas derived from source rocks in the overlying Xujiahe Formation (Qin Chuan et al., 2011). The gas reservoired in the 2nd member of the Upper Triassic Xujiahe Formation was widely believed to display characteristics of both oil-type and coal-type gases. However, Shen Zhongmin et al. (2009) considered that the gas was derived from its own respective set of source rocks, whereas Ye Jun (2001) proposed that the gas was derived from source rocks in the underlying Lower Triassic Ma'antang and Xiaotangzi formations. As for the natural gas reservoired in the Middle-Upper Jurassic strata, since the dark mudstone was hardly developed in western Sichuan Basin, and the Jurassic mudstone had a poor hydrocarbon potential due to the TOC content generally being lower than 0.5% (Cai Kaiping and Liao Shimeng, 2000), the Jurassic gas in western Sichuan Basin including the Xinchang gas field was believed to be derived from the underlying Xujiahe Formation coal-measure source rocks (Wu et al., 2010; Dai et al., 2012; Wu Xiaoqi et al., 2011). Although Shen Zhongmin et al. (2008) proposed that the Jurassic gas in the Xinchang gas field was derived from the 5th member of Xujiahe Formation source rocks based on the analysis of the rock association characteristics, there was still inadequate geochemical evidence of natural gas.

The divergence of the gas origin and source of the Xinchang gas field in the Sichuan Basin was mainly caused by the various understanding of the geochemical characteristics of natural gas, and previous studies on the gas geochemistry generally paid little attention to hydrogen isotopic composition. In this paper, we will systematically analyze the geochemical characteristics of natural gas based on the measurement of the chemical composition as well as stable carbon and hydrogen isotopic compositions. We will also further probe into the genetic types of natural gas in order to provide beneficial information for the gas-source correlation.

2 Geological Settings

The Sichuan Basin is an NE-trending and diamond tectonic-sedimentary basin in southwestern China, and is a large petroliferous basin superimposed by marine carbonate platform sediments and terrigenous clastic sediments covering an area of 19×10^4 km^2 (Tong Chongguang, 1992). The Sichuan Basin structurally belongs to the Yangtze Block, and it comprises a composite basin-mountain system with its peripheral mountains, which exerts significant control on the present distribution of medium-large sized natural gas fields in the basin (Liu et al., 2012).

The Western Sichuan Depression locates in the western Sichuan Basin, and thick T_3-K terrigenous strata were deposited as a result of the strike-slip and thrust faulting of the Longmenshan thrust belt in Late Triassic. The depression was commonly known as the Western Sichuan Foreland Basin (Deng et al., 2012; Liu et al., 2012).The Xinchang gas field is situated in the Xiaoquan–Xinchang–Hexingchang region in western Xinchang structural belt, which is in the central part of the Western Sichuan Depression. An NEE-trending anticline higher in the west and lower in the east was developed in this region. It has a steep south flank and a gentle north flank, and only a few thrust faults were developed in the eastern part of the region as seen from the upper surface of the Upper

Triassic Xujiahe Formation (Fig. 1).

The sequential drilled formations downwards are the Quaternay (Q), the Lower Cretaceous Jianmenguan Formation (K_1j), the Upper Jurassic Penglaizhen (J_3p) and Suining (J_3sn) formations, the Middle Jurassic Shaximiao (J_2s) and Qianfuya (J_2q) formations, the Lower Jurassic Baitianba Formation (J_1b), the Upper Triassic Xujiahe (T_3x), Xiaotangzi (T_3t) and Ma'antang (T_3m) formations, and the 4th member of the Middle Triassic Leikoupo Formation (T_2l^4) (Fig. 2). The Xujiahe Formation was generally divided into six members (T_3x^1–T_3x^6) from bottom to top in the Sichuan Basin (Dai et al., 2009), whereas only the 2nd to 5th members (T_3x^2–T_3x^5) were developed in the Xinchang gas field. The T_3x^6 is missing and J_1b uncomformably covers T_3x^5 as a result of denudation in the Xinchang gas field. Moreover, T_3t–T_3m corresponds to T_3x^1 in other regions of the Sichuan Basin. The Xinchang gas field is a large gas field with proven gas reserves of 204.522×10^9 m^3, and the natural gas is mainly reservoired in T_2l^4, T_3x^2, T_3x^4, T_3x^5, J_2q, J_2s and J_3p (Fig. 2). Natural gas in the Xinchang gas field is mainly reservoired in the Jurassic and Upper Triassic sandstones as well as the Middle Triassic limestone and dolomite (Fig. 2).

The Jurassic strata (J_1b, J_2q, J_2s, J_3s, and J_3p) in the Xinchang gas field are dominated by a set of inland lacustrine red clastic sediments with a thickness of 2000 m (Cai Kaiping and Liao Shimeng, 2000). They have low hydrocarbon generation potential due to the TOC contents generally lower than 0.2% (Dai et al., 2012). The Xujiahe Formation represents a set of lakeshore deposits and/or marsh facies with the thickness decreasing towards the southeast to the central Sichuan Basin (Dai et al., 2009). The dark mudstone has an average TOC content of 1.96%, not to mention, its interbedded coals are significant source rocks being both type II and III kerogen. They are mainly developed in T_3x^1, T_3x^3 and T_3x^5 in the Western Sichuan Depression (Dai et al., 2012). The Ma'antang and Xiaotangzi formations are a set of transitional facies deposited in shallow bay environment, and the marlstone and mudstone deposited in bay mudflat and shoreland marsh facies constitute source rocks with kerogen types I and II (Ye Jun, 2003). The Leikoupo Formation represents a set of deep lagoon facies of restricted- evaporate platforms. It mainly consists of limestone and dolomite with interbedded gypsum. The residual TOC contents of the carbonate rocks generally lie in 0.2%–0.4% (Xu Guoming et al., 2013). Nonetheless, whether the carbonate rocks could be considered source rocks is still controversial.

3 Samples and Methods

The gas samples from the Xinchang gas field in the Sichuan Basin were collected from the wellheads after flushing the lines for 15–20 minutes to remove air contamination. To collect the gas samples, 5 cm radius stainless steel cylinders with double valves were used.

The chemical composition of gas samples was determined using an Agilent 7890A Gas Chromatograph (GC) equipped with a flame ionization detector and a thermal conductivity detector. Individual alkane gas components were separated using a capillary column (PLOT Al_2O_3 50m×0.53mm). The GC oven temperature

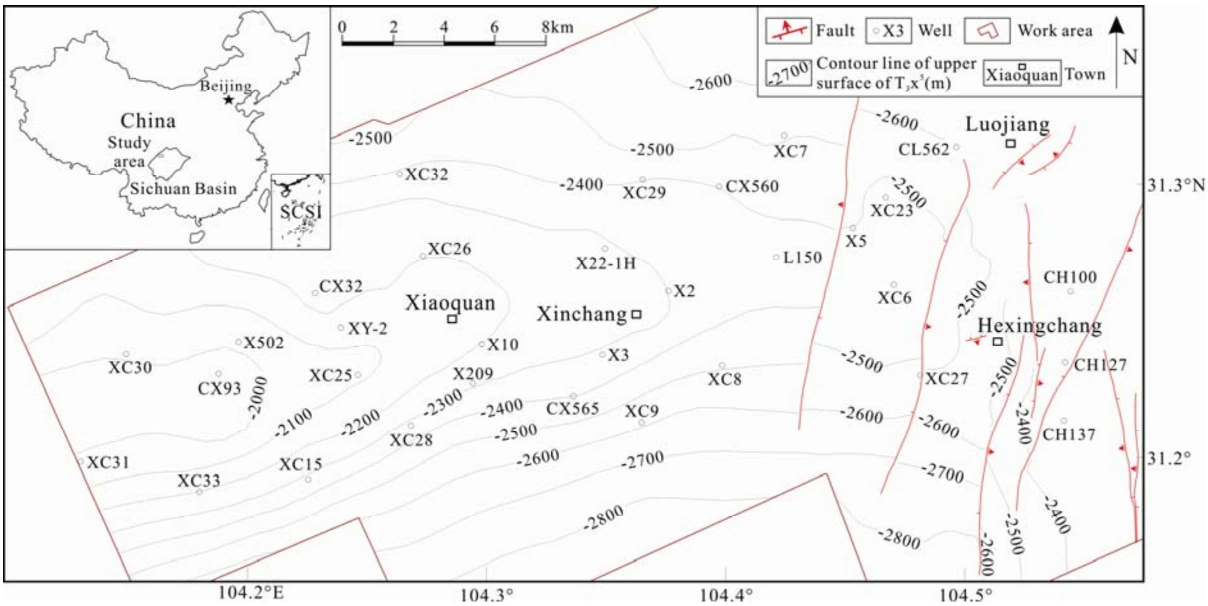

Fig. 1. Map showing location and main wells of the Xinchang gas field in western Sichuan Basin, southwestern China.

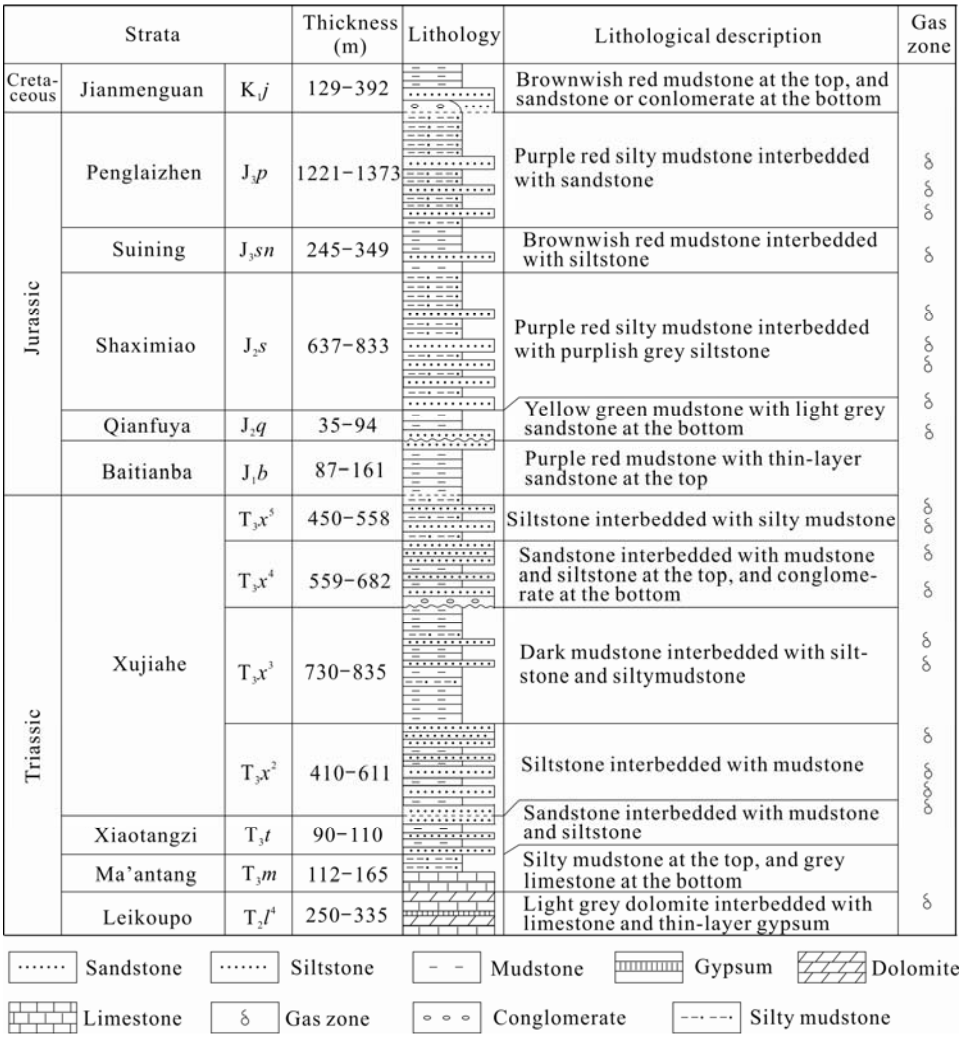

Fig. 2. Stratigraphic column of the Xinchang gas field in western Sichuan Basin.

was initially set at 40℃ for 5 minutes, heating at a rate of 10℃/min until it reached a final temperature of 180℃, which was held for 20 minutes,

Stable carbon isotopic composition of the natural gas was measured on a Finnigan MAT-253 mass spectrometer. The alkane gas components and CO_2 were initially separated using a fused silica capillary column (PLOT Q 30m×0.32mm) with helium as the carrier gas. The oven temperature was ramped from 40–180℃ at a heating rate of 10℃/min, and the final temperature was held for 10 minutes. Each gas sample was measured in triplicate. Stable carbon isotopic values are reported in the δ notation in permil (‰) relative to VPDB, and the measurement precision is estimated to be ±0.5‰ for δ^{13}C.

Stable hydrogen isotopic composition of alkane gases was measured on a Thermo Scientific Delta V Advantage mass spectrometer (GC/TC/IRMS). The alkane gas components were separated on an HP-PLOT Q column (30m×0.32mm×20μm) with helium as the carrier gas at 1.5ml/min. The GC oven was initially held at 30℃ for 5 minutes. It was then programmed to 80℃ at the heating rate of 8℃/min, and then it was subsequently heated to 260℃ at 4℃/min where it was held for 10 minutes. Each gas sample was measured in triplicate and the results were averaged. The measurement precision is estimated to be ±3‰ for δD with respect to VSMOW.

4 Results

The chemical compositions and stable carbon and hydrogen isotopic compositions of natural gas in the Xinchang gas field are listed in Table 1.

4.1 Chemical composition of natural gas

Natural gas in the Xinchang gas field within the Sichuan Basin is mainly composed of alkane gas with a low amount of nonhydrocarbons. The CH_4 content ranges between 88.99% and 98.01% with an average of 94.55%, and the C_2–C_5 content ranges in 0.33%–8.96% with an average of 4.22%. The CH_4 content is positively correlated

Table 1 Molecular composition and stable carbon and hydrogen isotopic compositions of natural gas in the Xinchang gas field

Well	Strata	Components (%)										δ^{13}C (‰), VPDB						δD (‰), V_{SMOW}			R_o(%)①	R_o(%)②	R_o(%)③
		CH_4	C_2H_6	C_3H_8	i-C_4	n-C_4	i-C_5	n-C_5	N_2	CO_2	H_2S	$\delta^{13}C_1$	$\delta^{13}C_2$	$\delta^{13}C_3$	$\delta^{13}iC_4$	$\delta^{13}nC_4$	$\delta^{13}C_{CO2}$	δD_1	δD_2	δD_3			
CH141	J_3p	95.41	3.16	0.76	0.14	0.15	0.05	0.03	0.23	0.06	0	−33.9	−24.4	−21.4	−19.9	−21.6	−14.1	−162	−138		1.08	3.35	1.74
CH358-1	J_3p	96.33	2.59	0.53	0.10	0.10	0.04	0.02	0.23	0.04	0	−34.5	−24.2	−21.9	−21.2	−20.2	−16.0	−165	−138		0.98	3.07	1.67
HP1	J_3p	93.84	3.61	0.90	0.15	0.19	0.07	0.06	0.85	0.23	0	−32.5	−23.6	−20.5		−19.3		−167	−128	−118	1.36	4.10	1.96
XQ111	J_3p	94.75	3.69	0.84	0.13	0.17	0.06	0.04	0.23	0.09	0	−35.0	−23.7	−22.2	−20.5	−22.5	−16.1	−169	−138		0.91	2.86	1.56
CX109	J_3p	92.46	4.27	1.03	0.20	0.24	0.09	0.07	1.36	0.07	0	−33.1	−24.8	−21.4				−180	−123	−121	1.23	3.75	1.84
HP16	J_3p	91.97	4.01	1.04	0.21	0.23	0.08	0.06	2.05	0.13	0	−32.9	−24.5	−21.9				−171	−139	−117	1.27	3.86	1.88
CX132	J_2s	96.58	2.48	0.42	0.08	0.08	0.03	0.00	0.24	0.07	0	−33.8	−22.6	−21.9	−20.6	−21.8	−16.3	−166	−142		1.10	3.40	1.78
CX170	J_2s	94.21	4.18	0.82	0.14	0.14	0.04	0.03	0.32	0.11	0	−35.0	−23.5	−20.8	−20.8	−21.4	−17.1	−171	−143		0.91	2.86	1.55
CX455	J_2s	94.36	3.18	0.75	0.15	0.16	0.07	0.07	0.96	0.09	0	−36.4	−24.6	−21.7		−24.6		−174	−133	−109	0.72	2.33	1.40
CX628	J_2s	93.59	4.28	1.00	0.20	0.19	0.08	0.04	0.54	0.05	0	−36.8	−25.4	−22.8	−22.2	−23.4	−15.4	−171	−144		0.68	2.20	1.32
CX454	J_2s	94.51	3.52	0.64	0.10	0.12	0.04	0.03	0.98	0	0	−35.0	−22.5	−19.1		−18.8					0.90	2.85	1.56
XS3-1HF	J_2s	95.07	3.16	0.63	0.12	0.13	0.05	0.03	0.59	0.07	0	−33.1	−24.0	−22.1				−173	−115		1.24	3.78	1.87
CX135	J_2q	92.73	4.42	1.09	0.26	0.24	0.10	0.07	0.89	0.20	0	−35.6	−23.5	−20.6	−20.5	−20.4	−15.8	−170	−137		0.82	2.62	1.48
CX152	J_2q	94.47	3.80	0.85	0.15	0.17	0.07	0.04	0.32	0.10	0	−35.4	−24.8	−22.2	−21.6	−23.6	−16.1	−170	−141		0.85	2.69	1.51
CX152	J_2q	94.43	2.43	0.60	0.20	0.14	0.06	0.04	1.53	0.32	0	−35.3	−25.2	−23.7		−25.2		−171	−133	−120	0.86	2.73	1.57
CX163	J_2q	94.86	2.97	0.55	0.12	0.10	0.04	0.03	0.68	0.58	0	−35.0	−23.3	−20.6							0.90	2.85	1.58
CX152	J_2q	93.97	3.52	0.81	0.15	0.17	0.06	0.04	1.04	0.06	0	−33.7	−25.0	−22.7				−172	−129		1.13	3.48	1.77
X502	T_3x^5	89.62	5.32	2.02	0.42	0.44	0.18	0.13	1.09	0.49	0	−36.1	−26.0	−22.2				−197	−138	−123	0.76	2.44	1.41
XC23	T_3x^5	89.93	5.86	1.71	0.36	0.33	0.14	0.09	0.76	0.62	0	−39.1	−26.0	−21.6			−11.7	−198	−136	−123	0.46	1.57	1.07
XC25	T_3x^5	92.38	4.21	1.31	0.25	0.27	0.11	0.08	0.67	0.51	0	−34.6	−24.9	−21.8				−182	−132		0.96	3.02	1.61
XC26	T_3x^5	88.99	6.23	1.79	0.36	0.34	0.14	0.09	1.09	0.70	0	−40.2	−25.8	−21.4			−6.2	−190	−126		0.39	1.33	0.97
XC30	T_3x^5	89.73	5.64	2.23	0.42	0.40	0.11	0.07	1.20	0.13	0	−37.9	−27.3	−23.2				−193	−139	−115	0.56	1.87	1.18
XYHF-2	T_3x^5	93.31	3.81	1.01	0.22	0.21	0.10	0.07	0.58	0.48	0	−34.9	−24.5	−21.2				−192	−131		0.92	2.89	1.58
CX560	T_3x^4	97.24	2.00	0.21	0.03	0.02	0.01	0.00	0.29	0.12	0	−33.7	−20.8	−20.6							1.12	3.44	1.81
X22	T_3x^4	95.00	3.58	0.36	0.07	0.05	0.04	0.02	0.35	0.53	0	−33.4	−21.4								1.18	3.63	1.81
X882	T_3x^4	95.80	2.55	0.56	0.13	0.12	0.05	0.03	0.20	0.52	0	−33.4	−22.1	−20.6		−19.9					1.17	3.59	1.84
X882	T_3x^4	95.73	2.50	0.49	0.10	0.10	0.05	0.03	0.68	0.25	0	−35.2	−24.3	−22.9		−24.3		−171	−137	−125	0.88	2.77	1.57
XC28	T_3x^4	94.35	4.17	0.41	0.07	0.05	0	0	0	0.96	0	−35.1	−20.4	−19.1			−8.9	−174			0.89	2.81	1.52
X22	T_3x^4	92.95	5.66	0.87	0.18	0.14	0.09	0.04	0.01	0	0	−33.4	−21.8				−4.9	−173	−131		1.18	3.61	1.74
X21-1H	T_3x^4	94.77	4.08	0.70	0.15	0.12	0.07	0.03	0.02	0	0	−31.4	−21.1				−0.4	−173	−119		1.63	4.83	2.15
CH127	T_3x^2	97.86	0.83	0.07	0.01	0.01	0.00	0.00	0.37	0.69	0	−30.7	−24.9	−25.5				−163	−161	−123	1.82	5.34	2.53
X5	T_3x^2	97.54	0.94	0.10	0	0	0	0	0	1.42	0	−31.6	−28.4	−28.2			−8.1	−166			1.58	4.69	2.30
X8-1H	T_3x^2	97.27	0.80	0.08	0	0	0	0	0.55	1.28	0	−32.2	−26.7	−25.5				−159	−148		1.43	4.29	2.21
X853	T_3x^2	97.51	0.81	0.08	0.01	0.01	0.00	0.00	0.27	1.28	0	−31.8	−26.9	−25.7			−1.8				1.53	4.57	2.30
X856	T_3x^2	97.10	0.70	0.07	0.01	0.01	0.00	0.00	0.62	1.40	0	−30.8	−27.0	−26.5			−0.8	−157	−151	−115	1.81	5.30	2.55
XC6	T_3x^2	97.39	1.06	0.12	0	0	0	0	0.00	1.43	0	−31.9	−26.4	−26.8			−7.5	−165			1.50	4.49	2.22
X2	T_3x^2	98.01	1.52	0.08	0.01	0.01			0.12	0.25	0	−31.9	−28.3								1.51	4.50	2.18
CK1	T_2l^4	94.57	0.31	0.02	0	0	0	0	0.30	4.78	0.02	−33.2	−34.8	−32.6			−3.7	−147	−131		1.21	3.71	2.33
XS1	T_2l^4	97.01	0.91	0.09	0	0	0	0	0.33	0.91	0.65	−33.6	−30.8	−29.0				−157			1.14	3.50	1.93

Note: Vitrinite reflectance values (R_o) were calculated according to different calibration equations for coal-type gas (①) and oil-type gas (②) proposed by Dai Jinxing (1992) and mixing gas (③) proposed by Pang Xiongqi et al. (2000).

with the dryness coefficient ($C_1/C_{1–5}$), which ranges in 0.908–0.997 with an average of 0.957. The gases in T_3x^5 are mainly wet with the dryness coefficient generally lower than 0.95, whereas those in T_2l^4 and T_3x^2 are typically dry with the dryness coefficient generally higher than 0.98. Natural gases in the Jurassic strata (J_2q, J_2s and J_3p) include both dry and wet gases with the dryness coefficient ranging in 0.92–0.98; the said situation is similar to those in T_3x^4 (Fig. 3a).

The nonhydrocarbon gases are mainly CO_2 and N_2, and their contents lie in 0–4.78% and 0–2.05% with an average of 0.54% and 0.58%, respectively (Table 1; Fig. 3b). A small amount of H_2S exists in the natural gas reservoired in the marine strata (T_2l^4), whereas no H_2S exists in the gases reservoired in the terrigenous strata (T_3x^2, T_3x^4, T_3x^5, J_2q, J_2s and J_3p) (Table 1).

4.2 Carbon isotopic composition of alkane gas

Natural gases reservoired in T_3x and the Jurassic strata within the Xinchang gas field display the $\delta^{13}C_1$, $\delta^{13}C_2$ and $\delta^{13}C_3$ values of −40.2‰ to −30.7‰, −28.4‰ to −20.4‰, and −28.2‰ to −19.1‰, respectively (Table 1; Fig. 4). Although the two $\delta^{13}C_1$ values (−33.6‰, −33.2‰) of gas samples from T_2l^4 are consistent with the $\delta^{13}C_1$ value range of gases in T_3x and the Jurassic strata, the $\delta^{13}C_2$ (−34.8‰, −30.8‰) and $\delta^{13}C_3$ (−32.6‰, −29.0‰) values are significantly lower than those in T_3x and the Jurassic strata (Table 1; Fig. 4).

The alkane gases in the Xinchang gas field mainly display positive carbon isotopic series, i.e., $\delta^{13}C_1<\delta^{13}C_2<\delta^{13}C_3$. However, two gas samples reservoired in T_3x^2 display partial reversal between ethane and propane ($\delta^{13}C_2>\delta^{13}C_3$), and one gas sample reservoired in T_2l^4 displays partial reversal between methane and ethane ($\delta^{13}C_1>\delta^{13}C_2$) (Fig. 4; Table 1).

4.3 Hydrogen isotopic composition of alkane gas

The δD_1 and δD_2 values of natural gases reservoired in T_3x and the Jurassic strata within the Xinchang gas field range from −198‰ to −157‰ and −161‰ to −115‰, respectively. The overall δD_1 values of gases reservoired in T_3x^5 are generally low and vary between −198‰ to −182‰. The δD_1 values (−157‰, −147‰) of gases reservoired in T_2l^4 are generally higher than those in T_3x and the Jurassic strata. The natural gases in the Xinchang gas field display positive hydrogen isotopic series with $\delta D_1<\delta D_2<\delta D_3$ (Table 1; Fig. 5).

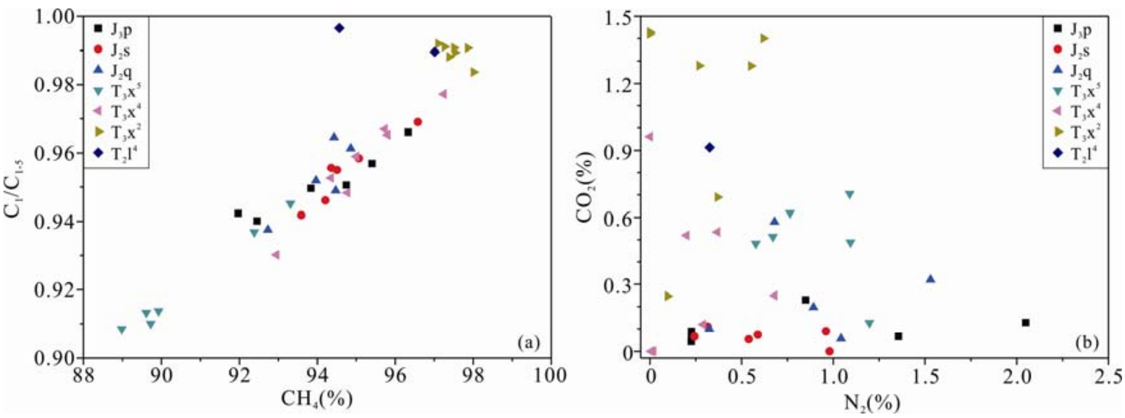

Fig. 3. Compositional diagrams of natural gas from the Xinchang gas field in western Sichuan Basin.
(a), C_1/C_{1-5} vs. CH_4; (b), CO_2 vs. N_2.

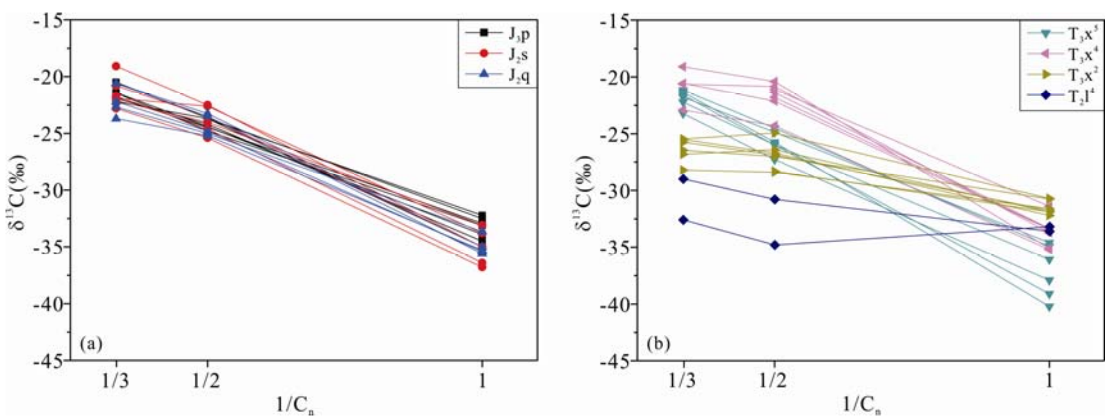

Fig. 4. Stable carbon isotopic series of alkane gases from the Xinchang gas field.
(a), Middle-Upper Jurassic; (b), Middle-Upper Triassic.

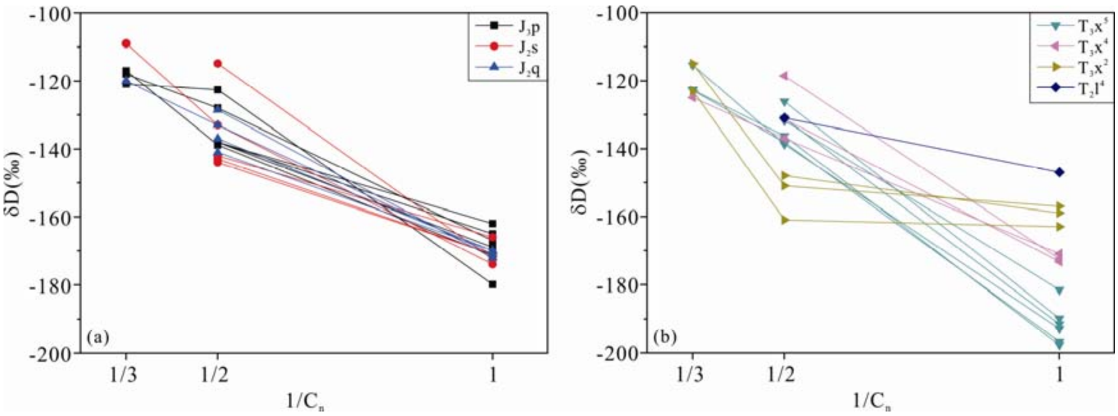

Fig. 5. Stable hydrogen isotopic series of alkane gases from the Xinchang gas field.
(a), Middle-Upper Jurassic; (b), Middle-Upper Triassic.

5 Discussions

5.1 Genetic types of alkane gas

Natural gas can be divided into biogenic and abiogenic gases according to their generation mechanism. Biogenic gas is derived from sedimentary organic matter, whereas the abiogenic gas is derived from inorganic matter (Dai Jinxing et al., 1992). The biogenic gas generally displays positive alkane carbon isotopic series with $\delta^{13}C_1$ values lower than −30‰, whereas the abiogenic gas displays negative series with $\delta^{13}C_1$ values higher than −30‰ (Dai Jinxing et al., 1992, 2008). The gas in the Xinchang gas field within the Sichuan Basin mainly displays positive alkane carbon isotopic series. Meanwhile, the $\delta^{13}C_1$ values are lower than −30‰ (Fig.4), indicative of typical characteristics of the biogenic gas.

The biogenic gas includes bacterial and thermogenic gas. The latter can be generally divided into coal-type gas derived from humic organic matter and oil-type gas derived from sapropelic organic matter (Dai Jinxing et al., 1992). The bacterial gas is primarily made up of methane with $\delta^{13}C_1$ values mainly lower than −55‰ and it displays different distribution characteristics with thermogenic gas being in the Bernard diagram (Bernard et al., 1976; Whiticar, 1999). Natural gases in the Xinchang gas field are significantly different from bacterial gas with $\delta^{13}C_1$ values higher than −55‰ (Fig. 4). Propane will be consumed preferentially during the biodegradation and its content will decrease. Therefore, natural gas affected by biodegradation will display rapidly increasing C_2/C_3 ratio with nearly constant C_2/iC_4 ratio, which is significantly different from the maturity trend (Prinzhofer and Battani, 2003). The alkane gases in the Xinchang gas field follow the maturity trend rather than the biodegradation trend in the correlation diagram between C_2/C_3 and C_2/iC_4 (Fig. 6).

Natural gases in the Xinchang gas field display typical characteristics of thermogenic gas rather than bacterial gas in the modified Bernard diagram (Fig. 7). The gases reservoired in T_3x^4, T_3x^5, and the Jurassic strata are generally consistent with the natural gas generated from type III kerogen, whereas those in T_2l^4 are similar to the range of natural gas derived from type II kerogen. Meanwhile, gases in T_3x^2 mainly display mixed or transitional characteristics of gases derived from types II

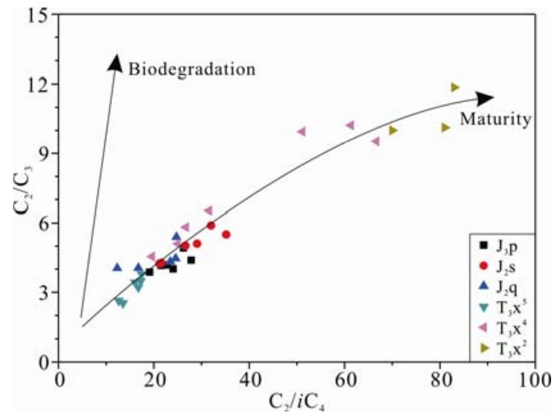

Fig. 6. Correlation diagrams between the C_2/C_3 and C_2/iC_4 values of natural gas from the Xinchang gas field.
(a), Middle-Upper Jurassic; (b), Middle-Upper Triassic (after Prinzhofer and Battani, 2003).

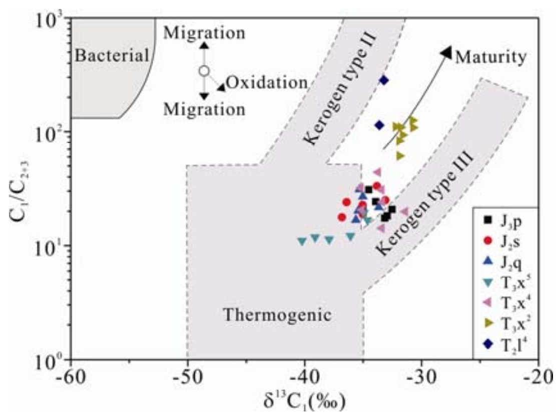

Fig. 7. Modified Bernard diagram of natural gas from the Xinchang gas field (after Bernard et al., 1976).

and III kerogens (Fig. 7).

The gases derived from different types of organic matter display different evolution trends in the correlation diagram between $\delta^{13}C_1$ and $\delta^{13}C_2$ values, and coal-type gas derived from humic organic matter has relatively higher $\delta^{13}C_2$ value than oil-type gas derived from sapropelic organic matter with similar $\delta^{13}C_1$ value (Rooney et al., 1995). In the correlation diagram between $\delta^{13}C_2$ vs. $\delta^{13}C_1$ (Fig. 8), the gases reservoired in T_3x^4, T_3x^5, and the Jurassic strata in the Xinchang gas field follow the trends of coal-type gases derived from type III kerogen in the Niger Delta (Rooney et al., 1995) and Sacramento Bain (Jenden et al., 1988), whereas those in the T_2l^4 are close to the trend of oil-type gas derived from type II kerogen in the Delaware/Val Verde Basin (Rooney et al., 1995), and the gases reservoired in the T_3x^2 display the transitional characteristics, indicating the occurrence of possible mixing between coal-type and oil-type gases.

The carbon isotopic compositions of alkane gases are affected by the thermal maturity of source rocks, and the $\delta^{13}C_1$, $\delta^{13}C_2$ and $\delta^{13}C_3$ values increase simultaneously with the thermal maturity (Dai Jinxing et al., 1992). However, the $\delta^{13}C_2$ value is far less affected by the maturity than the $\delta^{13}C_1$ value, and the carbon isotopes of ethane generally inherited those of organic matter in the source together with the $\delta^{13}C_2$ value is an effective index to identify coal-type and oil-type gases (Dai Jinxing et al., 2005). Empirical observations indicate that coal-type gas generally displays $\delta^{13}C_2$ and $\delta^{13}C_3$ values higher than −28‰ and −25‰, respectively, whereas the oil-type gas displays the opposite features (Liu et al., 2015). Natural gases reservoired in T_3x^4, T_3x^5, and the Jurassic strata display typical characteristics of coal-type gas with $\delta^{13}C_2$ and $\delta^{13}C_3$ values higher than −28‰ and −25‰, respectively. The $\delta^{13}C_2$ and $\delta^{13}C_3$ values are positively correlated, thus, indicating a maturity trend for coal-type gas (Fig. 9). Natural gases reservoired in T_2l^4 display a significantly higher thermal maturity than those in T_3x^4, T_3x^5, and the Jurassic strata in the modified Bernard diagram (Fig. 7). However, they display significantly lower $\delta^{13}C_2$ and $\delta^{13}C_3$ values than the latter. Therefore, they are typically oil-type gas with $\delta^{13}C_2$ and $\delta^{13}C_3$ values s lower than −28‰ and −25‰ (Fig. 9), which were evidently different from the T_3x^4, T_3x^5, and the Jurassic coal-type gas. The gases reservoired in T_3x^2 display significantly higher thermal maturity than those in T_3x^4, T_3x^5, and the Jurassic strata in terms of the correlation diagram between C_2/C_3 and C_2/iC_4 (Fig. 6) and the modified Bernard diagram (Fig. 7). They also display slightly lower thermal maturity than those in T_2l^4 in the modified Bernard diagram (Fig. 7). Nevertheless, if the gases reservoired in T_3x^2 were coal-type gases, they should display higher $\delta^{13}C_2$ and $\delta^{13}C_3$ values than those in T_3x^4, T_3x^5, and the Jurassic strata. If the T_3x^2 gases were typically oil-type gases, they should display lower $\delta^{13}C_2$ and $\delta^{13}C_3$ values than those in T_2l^4. However, the T_3x^2 gases display lower $\delta^{13}C_2$ and $\delta^{13}C_3$ values than T_3x^4, T_3x^5, and the Jurassic gases, but higher $\delta^{13}C_2$ and $\delta^{13}C_3$ values than T_2l^4 gase; these indicate that the T_3x^2 gases are different from typical T_3x^4, T_3x^5, and the Jurassic coal-type gases or T_2l^4 oil-type gases. They are located in the mixing area between oil-type and coal-types gases (Fig. 9). This is also consistent with the mixing trend of T_3x^2 gases indicated in the correlation diagram between $\delta^{13}C_2$ and $\delta^{13}C_1$ values (Fig. 8).

The hydrogen isotopic compositions of natural gas are controlled by the types of organic matter, thermal maturity and the environmental conditions of the aqueous medium, not to mention it could also be used to identify the gas origin (Dai Jinxing et al., 1992; Wang et al., 2015). The oil-type gases derived from sapropelic organic matter generally have less negative δD_1 values, whereas the coal-type gas derived from humic organic matter generally have more negative δD_1 values (Schoell, 1980; Wang et

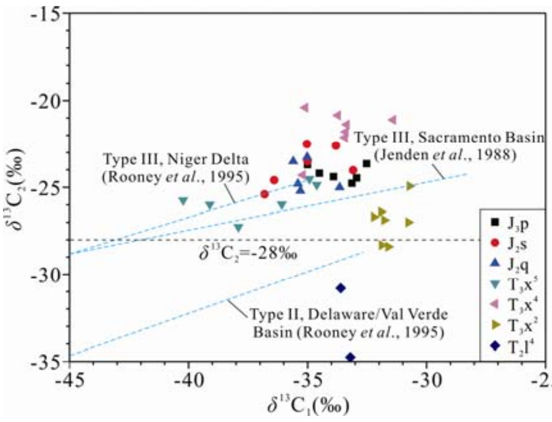

Fig. 8. Correlation diagram between the $\delta^{13}C_2$ and $\delta^{13}C_1$ values of natural gas from the Xinchang gas field.

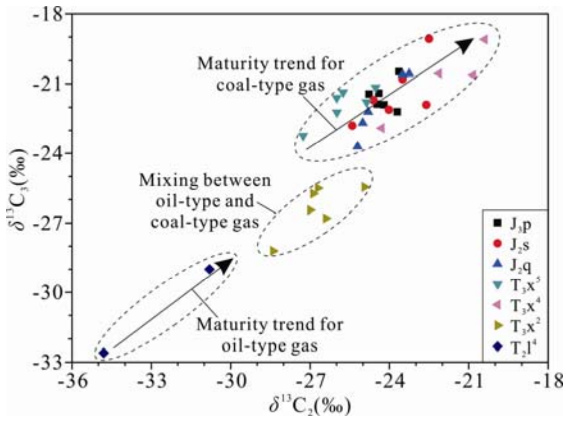

Fig. 9. Correlation diagram between the $\delta^{13}C_3$ and $\delta^{13}C_2$ values of natural gas from the Xinchang gas field.

al., 2015). The cutoff δD_1 value for marine and terrigenous natural gases in the Sichuan Basin was generally considered as −160‰ (Yu Cong et al., 2014). Natural gases reservoired in the T_2l^4 display the characteristics of marine gas with δD_1 values higher than −160‰ (Table 1), and are close to the trend of sapropelic gases from the Delaware/Val Verde Basin (Schoell, 1980) in the correlation diagram between δD_1 and $\delta^{13}C_1$ values (Fig. 10). Natural gases reservoired in T_3x^2, T_3x^4, T_3x^5, and the Jurassic strata in the Xinchang gas field display the characteristics of terrigenous gas with δD_1 values lower than −160‰ (Table 1). These gases follow the trend of coal gases from Northwestern Germany (Schoell, 1980) in Fig. 10. It is noteworthy that the T_3x^2 gases do not display the mixing trend between coal-type and oil-type gases in Fig. 10, indicating that the methane in T_3x^2 gases is mainly coal-type.

The statistical result of 313 natural gas samples from six Chinese sedimentary basins indicates that coal-type and oil-type gases display different distribution trends in the correlation diagram between δD_1 and $\delta^{13}C_2$ values (Wang et al., 2015). Natural gases reservoired in T_3x^4, T_3x^5, and the Jurassic strata in the Xinchang gas field are mainly consistent with the coal-type gases from humic organic matter in Fig. 11, whereas those in T_2l^4 are highly alike the oil-type gases from sapropelic organic matter. Furthermore, the gases reservoired in T_3x^2 are different from typical coal-type or oil-type gas since they follow the mixing trend between them in Fig. 11.

Prinzhofer and Pernaton (1997) proposed the correlation diagram between C_2/C_1 and $\delta^{13}C_1$ to distinguish the mixing, diffusion and maturity trend of natural gas. Natural gases reservoired in the T_3x^4, T_3x^5, and the Jurassic strata in the Xinchang gas field generally follow the maturity trend in the diagram of C_2/C_1 vs. $\delta^{13}C_1$ (Fig. 12), whereas the T_3x^2 gases follow the mixing with the oil-type gas trend.

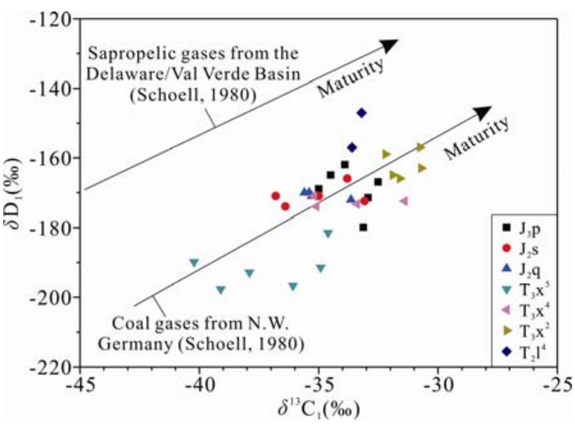

Fig. 10. Correlation diagram between the δD_1 and $\delta^{13}C_1$ values of natural gas from the Xinchang gas field.

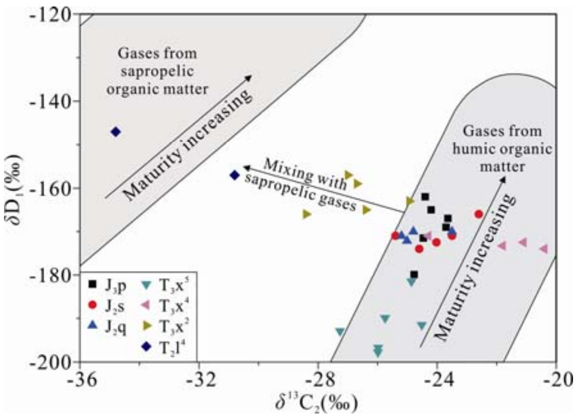

Fig. 11. Correlation diagram between the δD_1 and $\delta^{13}C_2$ values of natural gas from the Xinchang gas field (after Wang et al., 2015).

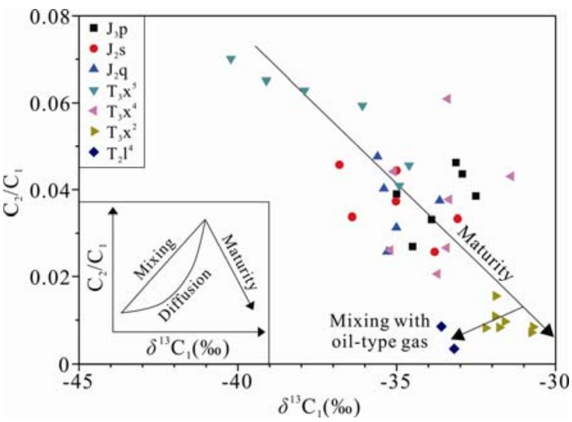

Fig. 12. Correlation diagram between the C_2/C_1 and $\delta^{13}C_1$ values of natural gas from the Xinchang gas field (after Prinzhofer and Pernaton, 1997).

Overall, the gases reservoired in T_3x^4, T_3x^5, and the Jurassic strata in the Xinchang gas field are coal-type gases and are derived from humic organic matter, whereas those in T_2l^4 are oil-type gases derived from sapropelic organic matter. The T_3x^2 gases have distinct $\delta^{13}C_2$ and $\delta^{13}C_3$ values in comparison with T_3x^4 and T_2l^4 gases of the Xinchang gas field in Fig. 9. In addition, they follow the coal-type gas trend in Fig. 10, which indicate that methane in T_3x^2 gases is mainly coal-type gas by means of ethane and propane being mixed together by coal-type and oil-type gases. Therefore, considering the high dryness coefficients (Fig. 3), the T_3x^2 gases are mainly coal-type gases assisted by a small amount of oil-type gas.

Oil-type gas can be generated from sapropelic organic matter through two approaches, the primary cracking of kerogen and secondary cracking of oil and / or gas. The primary and secondary cracking gases follow different variation trends of $\delta^{13}C_2$–$\delta^{13}C_3$ values and C_2/C_3 ratios with the increase of thermal maturity. Thus, the $\delta^{13}C_2$–$\delta^{13}C_3$ vs. C_2/C_3 correlation diagram could be used to identify the primary kerogen cracking gas and secondary oil/gas cracking gas (Prinzhofer and Huc, 1995; Lorant et al., 1998; Prinzhofer and Battani, 2003). The T_2l^4 gases in the Xinchang gas field have $\delta^{13}C_2$–$\delta^{13}C_3$ values of −2.2‰ and −1.8‰ with C_2/C_3 ratios of 15.50 and 10.12 (Table 1), respectively. These findings are consistent with the secondary oil cracking gas rather than the primary kerogen cracking gas in the $\delta^{13}C_2$–$\delta^{13}C_3$ vs. C_2/C_3 correlation diagram (Lorant et al., 1998). Consequently, the T_2l^4 gases in the Xinchang gas field are derived from the secondary cracking of oil.

5.2 Gas-source correlation

The potential source rocks revealed by the drilling wells in the Western Sichuan Depression consist of T_3x coal measures, T_3m and T_3t marlstone, mudstone, as well as T_2l carbonate rocks. The Upper Permian source rocks were not encountered during the drilling in the Western Sichuan Depression mainly due to the extremely large burial depth. The Upper Permian source rocks are a set of high-quality source rocks from the view of the entire Sichuan Basin, and are believed to be the main source for the giant marine gas reservoirs in the Upper Permian Changxing and Lower Triassic Feixianguan formations in northeastern Sichuan Basin (Hao et al., 2008).

The T_3x coal measures are the most important source rocks in the Western Sichuan Depression, and the $\delta^{13}C$ values of kerogen range from −26.5‰ to −24.0‰ with the kerogen type being II_2–III (Dai et al., 2009). The T_3x^2, T_3x^3, T_3x^4, and T_3x^5 source rocks display average TOC contents of 3.47%, 1.77%, 1.83%, and 2.35%, respectively, with average vitrinite reflectance (R_o) values of 2.47%, 2.06%, 1.69%, and 1.31%, correspondingly. These values indicate a stage of either mature to over-mature stage (Wang Dongyan et al., 2010).

The T_3m and T_3t marlstone and mudstone in the Western Sichuan Depression display kerogen type I–II, high TOC contents, and high thermal maturity (Ye Jun, 2003). For example, the T_3m dark mudstone in the Xinchang gas field displays an average TOC content of 1.13%, mean R_o value of 2.91%, and the average $\delta^{13}C$ values of kerogen in the T_3m and T_3t source rocks that range from −25.5‰ to −25.0‰, suggesting sapropelic-humic organic type (Wang Dongyan et al., 2010).

The T_2l carbonate rocks in the Xinchang gas field display kerogen type index (TI) that varies in 12.5%–98.03% with the kerogen type being II_1–II_2. They are in the over-mature stage with the equivalent R_o values higher than 2.0% and low residual TOC contents. An example of this manifestation are the carbonate rocks from the wells Chuanke 1 and Xinshen 1, they display average TOC contents of 0.25% and 0.27%, respectively (Yang Keming, 2016). These values have not reached the lower limit of TOC (0.5%) for the effective carbonate source rocks (Zhang Shuichang et al., 2002). Therefore, the T_2l carbonate rocks probably display limited hydrocarbon potential.

The coal-type gases reservoired in T_3x and the Jurassic strata in the Xinchang gas field have been considered to be derived from coal-measure source rocks in T_3x (Wu et al., 2010; Dai et al., 2012). The T_3x source rocks were mainly deposited in terrigenous environment except for the T_3x^1 in marine-terrigenous facies (Zhu Rukai et al., 2009). The maceral of the coal-measure source rocks mainly consists of vitrinite (Wang Dongyan et al., 2010; Zhang Min et al., 2012). In addition, the kerogen in different members of T_3x displays similar $\delta^{13}C$ values with humic organic matter (Dai et al., 2009). The gases generated by different members of T_3x have similar geochemical characteristics which were difficult to differentiate. However, for the source rocks in the various members of T_3x in the same area such as the Xinchang gas field, the most evident difference is the thermal maturity caused by different burial depth and the geochemical characteristics. Geochemical characteristics such as the methane $\delta^{13}C$ value have been considered to reflect the thermal evolution extent of source rocks. Therefore, the comparison between the vitrinite reflectance (R_o) of source rocks in different members of T_3x and the thermal maturity estimated from the methane $\delta^{13}C$ value are an important approach to carrying out the gas-source correlation.

Various models have been developed to estimate the thermal maturity of source rocks based on the isotopic compositions of associated natural gas (Stahl and Carey Jr, 1975; Schoell, 1980; Shen Ping et al., 1987; Dai Jinxing and Qi Houfa, 1989; Tang et al., 2000). These models are proposed based on coal-type gas or oil-type gas and are unsuitable for mixed gas or gas derived from organic matter in transitional facies. Dai Jinxing (1992) proposed the empirical $\delta^{13}C_1$–R_o equations for coal-type and oil-type gases based on gas samples from Chinese petroliferous basins, which have been widely and effectively used in China. Pang Xiongqi et al. (2000) proposed the quantitative identification model of the evolution degree of natural gas from mixing source material, which could be used to estimate thermal maturity of source rocks for mixing gas.

The geochemical characteristics of saturated and aromatic hydrocarbons from source rocks provide understandable evidence for transgressions that occurred during the Upper Triassic Xujiahe stage in the Sichuan Basin with a great impact on the source input and depositional environment, and they are different from those of the regular swamp facies humic source rocks

(Zhang Min et al., 2012). Although the T_3x alkane gases in the Xinchang gas field mainly display the geochemical characteristics of the coal-type gas, the quantitative identification model of the evolution degree of mixing natural gas rather than the coal-type gas equation will be suitable to estimate thermal maturity of source rocks. The thermal maturity data calculated from the $\delta^{13}C_1$-R_o equations for the coal-type and oil-type gases (Dai Jinxing, 1992) as well as the quantitative identification model for mixing natural gas (Pang Xiongqi et al., 2000) are listed in Table 1.

The average measured vitrinite reflectance values for T_3x^2, T_3x^3, T_3x^4 and T_3x^5 mudstones are 2.47%, 2.06%, 1.69%, and 1.31%, respectively (Wang Dongyan et al., 2010). The estimated thermal maturity values based on the $\delta^{13}C_1$–R_o equations for the coal-type and oil-type gases (Dai Jinxing, 1992) are significantly lower and higher than the measured R_o values, respectively. However, the estimated thermal maturity values based on the quantitative identification model for mixing natural gas (Pang et al., 2000) combined with the parameters in the equations proposed by Dai Jinxing (1992) are consistent with the measured values. For example, the estimated R_o values for T_3x^5 natural gas range from 0.97%–1.61% with an average of 1.30%, which are well consistent with the measured R_o values for T_3x^5 source rocks (1.22%–1.45% with an average of 1.31%) (Wang Dongyan et al., 2010) and significantly lower than those for T_3x^4 source rocks (1.63%–1.83% with an average of 1.69%) (Wang Dongyan et al., 2010). Therefore, the comparison indicates that the T_3x^5 natural gas is derived from the source rocks in T_3x^5 itself.

The natural gas reservoired in the Jurassic stratain the western Sichuan Basin including the Xinchang gas field was believed to be derived from the underlying Xujiahe Formation coal-measure source rocks (Wu et al., 2010; Dai et al., 2012) due to insufficient hydrocarbon potential for the Jurassic strata (Cai Kaiping and Liao Shimeng, 2000), Although the Jurassic-reservoired gases in the Xinchang gas field had been considered to be derived from T_3x^5 source rocks (Shen Zhongmin et al., 2008), they display similar geochemical characteristics for dryness coefficient, methane carbon and hydrogen isotopic compositions with that of the T_3x^4 gases. They are also greatly different from the T_3x^5 gases. The average estimated R_o values for Jurassic and T_3x^4 gases are 1.65% and 1.78%, respectively. These values are similar to the average measured R_o values for T_3x^4 (1.69%) and T_3x^3 (2.06%) source rocks (Wang Dongyan et al., 2010), and they are significantly higher than the average measured R_o values for T_3x^5 source rocks (1.31%) (Wang Dongyan et al., 2010). Hence, the Jurassic and T_3x^4 gases display good affinity and are mainly derived from T_3x^3 and T_3x^4 source rocks.

The estimated R_o values for T_3x^2 gases range between 2.18%–2.55% with an average of 2.33%. These values are generally consistent with the measured R_o values for T_3x^2 source rocks (2.31%–2.57% with an average of 2.47%) (Wang Dongyan et al., 2010). They are generally higher and lower than the measured R_o values for T_3x^3 (2.06% on average) and T_3m (2.91% on average) source rocks (Wang et al., 2010), respectively. Since the T_3x^2 gases are mainly coal-type gas accompanied by a small amount of oil-type gas, and the source rocks in the T_3t and T_3m display kerogen types I and II (Ye Jun, 2003), the T_3x^2 gases are mainly derived from the source rocks in T_3x^2 itself with a small amount contributed by T_3t and T_3m source rocks.

Since the T_2l^4 gases are oil-type gases, the $\delta^{13}C_1$–R_o equation for the oil-type gases (Dai Jinxing, 1992) can be used to estimate the thermal maturity of the source rocks. The estimated thermal maturity for the two gas samples is 3.50% and 3.71% (Table 1), respectively. They are significantly higher than the R_o values of T_3m source rocks (2.91% on average) (Wang Dongyan et al., 2010). The T_2l and Lower Triassic strata were hardly considered as effective source rocks due to the extremely low TOC contents (generally lower than 0.5%), whereas the Upper Permian mudstone source rocks in the Sichuan Basin have been considered important (Hao et al., 2008). Therefore, the T_2l^4 gases are believed to be derived from the secondary cracking of oil that's most likely generated from the Upper Permian source rocks.

6 Conclusions

(1) The chemical compositions as well as carbon and hydrogen isotopic values for natural gas from the Xinchang gas field within the Sichuan Basin indicate that the natural gas is mainly composed of methane. The methane has a content ranging in 88.99%–98.01% with a dryness coefficient varying in 0.908–0.997. It generally displays positive alkane carbon and hydrogen isotopic series.

(2) The gases reservoired in T_3x^4, T_3x^5, and the Jurassic strata in the Xinchang gas field display $\delta^{13}C_2$ values higher than −28‰ and δD_1 values lower than −160‰, and are identified as typical coal-type gases. In addition, the gases reservoired in T_2l^4 are oil-type gases with $\delta^{13}C_2$ values lower than −28‰ and δD_1 values higher than −160‰. The T_3x^2 gases display transitional characteristics between these two types of gases and are mainly coal-type gas accompanied by a small amount of oil-type gas.

(3) The gas-source correlation indicates that the T_3x^5 natural gas is derived from the source rocks in T_3x^5 itself,

and the Jurassic and T_3x^4 gases display good affinity and are mainly derived from T_3x^3 and T_3x^4 source rocks. The T_3x^2 gases are mainly derived from the source rocks in T_3x^2 itself with a small amount of contribution by T_3t and T_3m source rocks, whereas the T_2l^4 gases are believed to be derived from the secondary cracking of oil most likely generated from the Upper Permian source rocks.

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