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Geochemical characteristics and origin of gases from the Upper, Lower Paleozoic and the Mesozoic reservoirs in the Ordos Basin, China

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The Ordos Basin, the second largest sedimentary basin in China, contains the broad distribution of natural gas types. So far, several giant gas fields have been discovered in the Upper and Lower Paleozoic in this basin, each having over 1000×10^8m ^{3} of proven gas reserves, and several gas pools have also been discovered in the Mesozoic. This paper collected the data of natural gases and elucidated the geochemical characteristics of gases from different reservoirs, and then discussed their origin. For hydrocarbons preserved in the Upper Paleozoic, the elevated δ^{13}C values of methane, ethane and propane indicate that the gases would be mainly coal-formed gases; the singular reversal in the stable carbon isotopes of gaseous alkanes suggests the mixed gases from humic sources with different maturity. In the Lower Paleozoic, the $\delta^{13}C_1$ values are mostly similar with those in the Upper Paleozoic, but the $\delta^{13}C_2$ and $\delta^{13}C_3$ values are slightly lighter, suggesting that the gases would be mixing of coal-type gases as a main member and oil-type gases. There are multiple reversals in carbon isotopes for gaseous alkanes, especially abnormal reversal for methane and ethane (i.e. $\delta^{13}C_1>\delta^{13}C_2$), inferring that gases would be mixed between high-mature coal-formed gases and oil-type gases. In the Mesozoic, the δ^{13}C values for gaseous alkanes are enriched in ^{12}C, indicating that the gases are mainly derived from sapropelic sources; the carbon isotopic reversal for propane and butane in the Mesozoic is caused by microbial oxidation and mixing of gases from sapropelic sources with different maturity. In contrast to the Upper Paleozoic gases, the Mesozoic gases are characterized by heavier carbon isotopes of iso-butane than normal butane, which may be caused by gases generated from different kerogen types. Finally, according to $\delta^{13}C_1$-R_o relationship and extremely low total organic carbon contents, the Low Paleozoic gases would not be generated from the Ordovician source as a main gas source, bycontrast, the Upper Paleozoic source as a main gas source is contributed to the Lower Paleozoic gases.

Ordos Basin, natural gas, chemical composition, carbon isotope, genesis

The Ordos Basin, located in the middle of China, is the second largest sedimentary basin and one of the most tectonically stable basins in China[1]. Several giant gas fields, each containing over 1000×10^8m^3 (bcm) of proven gas reserves, have been discovered in this basin, e.g. Sulige gas field as the largest gas field in China.

The natural gas in the Ordos Basin is characterized by widespread, various types and multiple preservation, so that many scientists have carried on much research into gas sources, gas types and regional distribution. But

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there are still in debate on the origin of Ordovician weathered crust gas[2−12]. Some thought the gas was derived dominantly from the carbonates of Ordovician Majiagou Formation, but others proposed that the gas was derived mainly from the Carboniferous-Permian coal measures. The divergence of opinions about the origin of the Lower Paleozoic gas would bring about a puzzle on carbonate evaluation, gas field discovery and exploration. Moreover, less attention has been paid to the Mesozoic gas. In order to figure out disputes and problems, the paper collected the data of the chemical and carbon isotopic compositions of gas from this basin and determined gas sources.

1 Introduction

1.1 Geological background

The Ordos Basin is a multicycle craton petroliferous basin with stable subsidence, depressional movement and torque (Figure 1)[1]. The Ordos Basin as one of the most tectonically stable basins in China is characterized by the whole ascending, durative subsidence, aclinic line and conformable contact[1]. The gas fields occur in the northern part of the basin, with relatively shallow Paleozoic gas sources and reservoir rocks. By contrast, several oil fields are distributed mainly in the southern part of the basin, with much greater burial depth. The

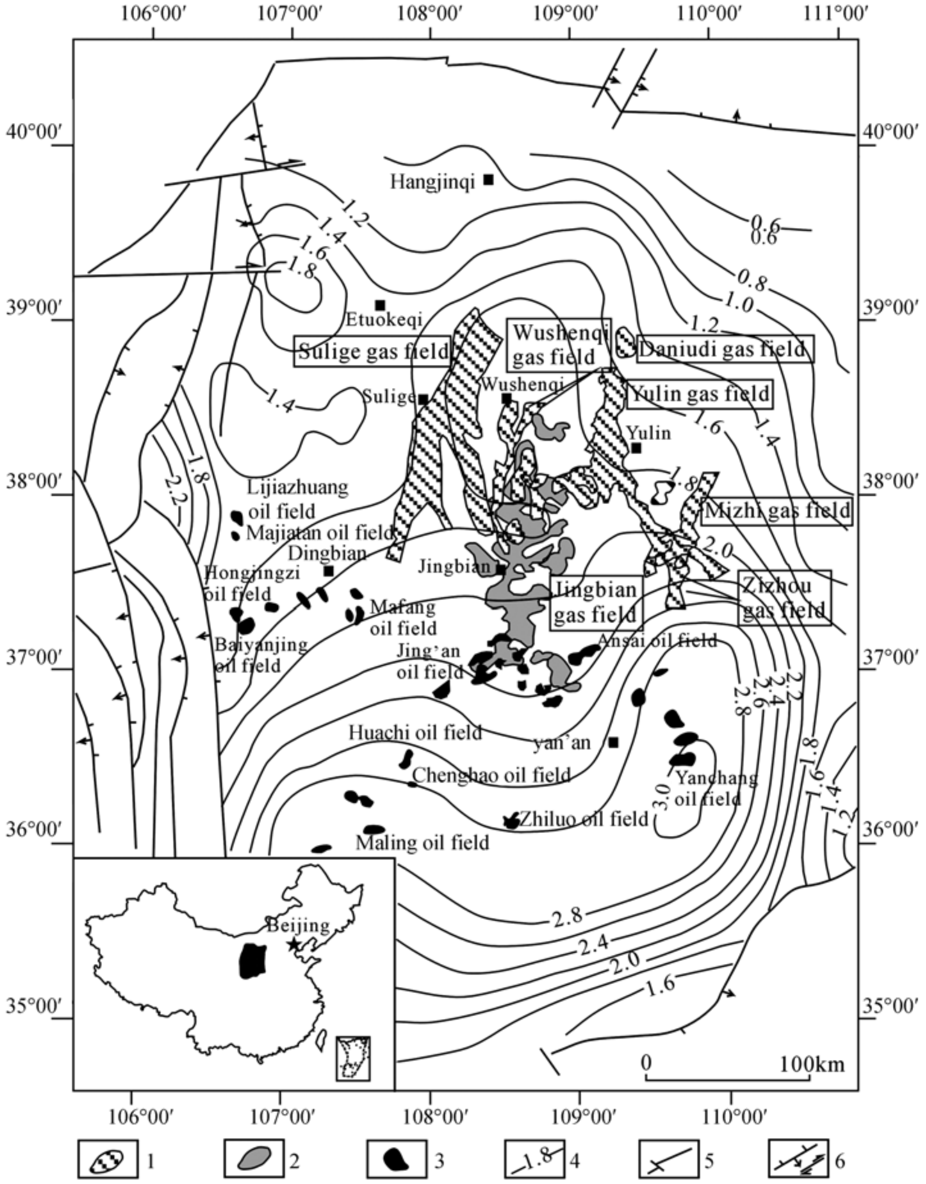

Figure 1 Map showing the oil and gas fields locations in the Ordos Basin. 1, Gas fields with Upper Paleozoic reservoirs; 2, gas fields with Lower Paleozoic reservoirs; 3, oil fields; 4, vitrinite reflectance isopleths (%R_o) at the base of the Carboniferous; 5, basin boundary; 6, fault.

possible sources for the oil lie within the Mesozoic strata[1]. Due to few structural reservoirs[13], most hydrocarbons are discovered in the lithologic traps[3].

1.2 Introduction of gas fields

The Sulige, Yulin, Wushenqi, Zizhou and Daniudi fields were developed in the Upper Paleozoic and Jingbian field was in the Lower Paleozoic in the Ordos Basin. These gas fields' relevant gas payzones include the Upper Permian Shihezi Formation (P_2sh); Lower Permian Xiashihezi (P_1x), Shanxi (P_1s) and Taiyuan Formation (P_1t); Carboniferous Benxi Formation (Cb) and Lower Ordovician Majiagou Formation (O_1m). The gases in the Sulige and Yulin fields are preserved in lithologic traps. The gases in the Wushenqi field are trapped in lithostructural trap, and the southern edge of this field is overlapped geographically with the northwestern portion of the Jingbian field. The Jingbian field consists of five gas pools, all in lithologic traps. The top gas pool is sealed by Carboniferous bauxitic mudstones and coal measure, whereas the others are capped by argillaceous dolomite and marlstone.

In the Mesozoic, the Lower Jurassic Yan'an Formation and the Upper Triassic Yanchang Formation are widely explored. Up to now, a few gas pools have been discovered, such as Zhiluo, Majiatan, Lizhuangzi, etc.[13]. The gas in Mesozoic reservoirs is widely distributed from the Upper Triassic Yanchang Formation (Chang10, Chang8, Chang4+5, Chang3, Chang2, Chang1) to the Lower Jurassic Yan'an Formation (Yan10, Yan9, Y8+7, Yan 5), as well as the Middle Jurassic Zhiluo Formation[13].

1.3 Source rocks

The dual sedimentary sequences are present in the Paleozoic strata of the Ordos Basin: an Upper Paleozoic sequence containing terrestrial clastics and coals, as well as alternate marine terrigenous sediments in the lower section; Lower Paleozoic containing marine carbonates and gypsum-salt sediments[1].

Two sedimentary sequences in the Upper Paleozoic have been considered as likely gas sources, including the Carboniferous-Permian coal measures and carbonates. The coal measures are the primary sources and distributed in the whole basin. The cumulative thickness of coal seams ranges from 10 to15 m over large area, with maximum of 40 m in some place and average TOC content is 60%[14]. The thickness of dark mudstones is mostly between 70—130 m, but occasionally up to 200 m. Exception for carbargilite, the C-P dark mudstones contain TOC content of 2%—4% as III kerogen[14]. The Carboniferous-Permian carbonates are the secondary source in the Upper Paleozoic and mainly distributed in the middle and east of the Basin. The TOC contents range from 0.3% to 5%, with average of 1.42%. The kerogen type of carbonates is humic-sapropelic type[12]. Additionally, the thermal maturity of organic matter in C-P is high stage (R_o ranges from 1.2% to 2.2% in the gas field areas of the basin and the maximum value is 2.8% in the southern part)[3] and the large total gas production and expulsion in the C-P is observed[15], which are the support for the giant gas fields in the Ordos Basin.

The geochemical characteristics of the Lower Paleozoic Ordovician carbonates are as follows: accumulative thickness of 400—500 m, TOC contents ranging from 0.08% to 0.64% with average of 0.22%[16], the sapropelic kerogan type, and R_o values between 2.07%—2.68%[14]. Although the Lower Paleozoic sources are of huge thickness and high maturity, there is no consensus on the Ordovician carbonates as a main source for the giant gas accumulations in the Lower Paleozoic because of the low TOC contents and dispersive hydrocarbon generation.

The dark mudstones with deep and semideep laustrine facies in the Triassic Yanchang Formation are sources in the Mesozoic, containing two types of keogen: humic kerogen including strata beyond Chang3 Formation in the top and sapropelic type kerogen from Chang8 Formation to Chang4+5 Formation in the middle[14], especially Chang7 Formation as the main source rock in the Mesozoic [14].

2 Component characteristics of natural gas in the Ordos Basin

Based on the gas components of 125 gas samples from the Upper Paleozoic reservoirs, 66 gas samples from the Lower Paleozoic reservoirs and 47 gas samples from the Mesozoic reservoirs (some are listed in Table 1), the frequency charts are shown in Figures 2 and 3.

2.1 Gaseous alkane components

Statistically, all of the gases from the Upper and Lower Paleozoic reservoirs are dominated by hydrocarbon gases, with high content of methane (Table 1, Figure 2).

Table 1 Main components and carbon isotopic compositions of gases from the Upper Paleozoic, the Lower Paleozoic and the Mesozoic reservoirs, the Ordos Basin (Part)

Gas field		Well	Strata	Gas main components (%)							Dryness	δ^{13}C (PDB, ‰)				
				CH_4	C_2H_6	C_3H_8	*i*-C_4	*n*-C_4	CO_2	N_2		$\delta^{13}C_1$	$\delta^{13}C_2$	$\delta^{13}C_3$	$\delta^{13}iC_4$	$\delta^{13}nC_4$
Upper Paleozoic	Sulige gas field	Su1	P_1s	91.57	4.52	0.89	0.19	0.16	0.78	0.71	0.941	−34.37	−22.13	−21.77	−21.53	−21.63
		Su6	P_1x	88.81	5.83	1.26	0.20	0.22	2.64	0.80	0.922	−33.54	−24.02	−24.72	−22.78	−23.23
		Su14	P_1x	96.37	1.66	0.40	0.13	0.09	1.25	0.00	0.977	−32.54	−23.17	−23.77		
		Su33-18	P_1x	91.69	4.26	0.91	0.25	0.29	0.87	1.43	0.941	−32.31	−25.23	−23.79	−22.20	−23.08
		Su36-13	P_1x	89.49	5.41	1.16	0.22	0.25	0.76	0.93	0.927	−33.40	−24.70	−24.40	−22.10	−23.10
		Tao5	P_1x	91.75	5.11	0.92	0.12	0.14	0.77	1.05	0.936	−33.05	−23.57	−23.72	−21.62	−22.46
		Tao6	P_1x	93.40	2.76	0.36	0.04	0.46	0.57	2.27	0.963	−29.00	−25.00	−27.00	−23.90	−25.70
	Yulin gas field	Shan141	P_1s	94.12	3.40	0.50	0.06	0.07	1.14	0.61	0.959	−33.7	−26.3	−24.30	−23.10	−23.10
		Shan143	P_1s	93.47	3.90	0.63	0.09	0.10	1.17	0.35	0.952	−33.57	−25.98	−24.42		−24.99
		Yu28-12	P_1s	94.25	3.25	0.47	0.06	0.07	1.08	0.65	0.961	−32.40	−27.00	−24.80	−23.60	−23.80
		Yu35-8	P_1s	95.32	2.67	0.34	0.07	0.06	1.45	0.03	0.968	−32.55	−24.87	−23.69	−21.17	−22.53
		Yu43-10	P_1s	94.39	2.73	0.41	0.08		1.80	0.52	0.967	−31.90	−26.40	−23.00	−23.69	−24.06
		Yu44-7	P_1s	95.65	2.65	0.32	0.04	0.04	0.57	0.69	0.969	−32.80	−25.50	−23.80	−23.80	−24.10
	Wushenqi Gas field	Shan215	P_1x	93.60	3.79	0.55	0.08	0.08	0.76	0.46	0.945	−32.90	−26.00	−24.00	−21.60	−22.90
		Shan241	P_1x	92.70	3.99	0.68	0.11	0.11	0.49	1.73	0.950	−32.60	−24.10	−24.20	−22.10	−23.30
		Shan243	P_1x	90.85	5.46	1.03	0.18	0.17	0.54	1.55	0.930	−35.00	−24.00	−23.60	−22.00	−22.90
		Wu22-7	P_1x	92.97	4.27	0.76	0.11	0.11	0.74	0.87	0.947	−32.60	−23.70	−24.20	−21.20	−22.70
		Yu19-5	P_1x	92.79	3.90	0.64	0.10	0.09	0.50	1.77	0.951	−34.50	−23.70	−24.70	−21.40	−23.50
		Yu24-5	P_1x	92.77	4.21	0.63	0.09	0.09	0.64	1.43	0.948	−32.20	−23.50	−24.90	−21.80	−23.60
Lower Paleozoic		Shan6	$O_1m_5^{1-4}$	92.60	0.32	0.03	0.00	0.00	4.86	2.22	0.996	−33.87	−34.05	−24.39		
		Shan12	$O_1m_5^{1-4}$	96.79	0.78	0.10	0.01	0.01	1.65	0.63	0.991	−34.21	−25.46	−26.37	−20.67	
		Shan15	$O_1m_5^{1-4}$	96.37	0.24	0.01			1.96	1.42	0.997	−33.19	−33.31	−25.87		
		Shan20	$O_1m_5^{1-3}$	93.10	0.30	0.16	0.01	0.02	0.55	1.02	0.995	−34.58	−30.96	−27.50	−22.10	
		Shan21	$O_1m_5^{1}$	95.92	1.33	0.17	0.02	0.03	2.36	0.11	0.984	−35.01	−24.59	−26.11	−24.27	
		Shan26	$O_1m_5^{3-4}$	91.07	0.15				3.04	5.53	0.998	−38.27	−34.13	−21.56	−25.17	
		Shan30	$O_1m_5^{4}$	86.07	0.77	0.10	0.00	0.00	0.41	12.01	0.990	−33.06	−33.58	−26.46		
		Shan34	$O_1m_5^{4}$	97.90	0.82	0.06	0.02	0.01	0.69	0.47	0.991	−33.99	−24.51	−22.42		
		Shan41	$O_1m_5^{1-7}$	98.14	1.51	0.20	0.07	0.04			0.982	−38.87	−28.67	−22.62	−20.40	
		Shan79	$O_1m_5^{5}$	96.55	0.08				0.45	2.79	0.999	−37.25	−31.81			
		Shan84	O_1m_5	92.40	0.81	0.12	0.01	0.01	5.09	0.99	0.990	−31.77	−28.49	−24.24		
		Shan106	$O_1m_5^{1}$	98.29	0.22	0.03			0.37	1.06	0.998	−30.66	−37.53	−29.95		
		E1	O_1m_5	90.70	7.05	1.50	0.47	0.17			0.908	−40.50	−31.40	−24.83		
		E5	O_1m_5	93.45	5.71				0.72	0.09	0.942	−41.74	−28.76			
		Futan1	O_1m_5	91.59	0.39	0.06			5.59	2.38	0.995	−33.74	−37.49	−18.20		
		Lin2	O_1m	94.25	1.31	0.19	0.02	0.02	1.51	2.62	0.984	−35.55	−25.57	−25.03		
Mesozoic		Liu131-12	T_3y	60.15	13.36	16.69	1.86	5.33	0.12		0.606	−36.67	−35.25	−32.41	−34.26	−31.62
		Liu133-10	T_3y	58.64	11.96	15.52	1.83	5.75	0.44	2.53	0.610	−42.30	−37.48	−33.51	−35.36	−32.37
		Liu94-35	T_3y	56.81	12.97	17.81	1.98	5.20	0.05	2.86	0.589	−43.65	−37.16	−33.48	−34.60	−32.64
		Ding31-7	J_1y	51.11	6.00	9.13	3.78	6.40	1.07	15.97	0.669	−41.50	−35.00	−32.10	−31.50	−31.40
		Sai1	T_3y	46.46	8.19	12.34	2.44	3.87	0.64	24.03	0.634	−52.57	−33.77	−32.38	−32.81	
		Sai34	T_3y	80.04	7.64	5.57	0.61	1.45	0.17	3.28	0.840	−49.46	−37.58	−33.65	−32.91	
		Quan36	J_1y	92.50	2.34	0.32	0.15	0.05	0.10	4.49	0.970	−59.69	−30.31	−25.57	−31.88	
		Ma254	T_3y	98.08	0.34				0.36	1.22	0.997	−48.00				
		Ma9-1	J_1y	95.47	0.86	0.60	0.24	0.44	0.12	1.63	0.978	−47.20	−33.70	−28.80	−31.40	−30.40
		Hu401	T_3y	91.53	1.55	0.60	1.59	1.12	0.84	2.40	0.950	−48.20	−30.78	−30.16	−31.26	
		Hu43-10	T_3y	57.51	3.88	6.01	2.33	1.86	0.32	20.38	0.803	−45.10	−35.60	−33.20	−32.80	−32.70
		Cheng9-28	J_1y	65.21	12.75	10.28	1.34	2.24	1.43	5.85	0.710	−47.46	−36.29	−32.70	−31.64	
		Luo35-34	T_3y	38.14	10.76	15.56	3.37	10.18	0.18	10.80	0.489	−48.00	−38.50	−33.70		−32.40
		Geng20	J_1y	60.08	13.75	16.34	1.83	3.96	173		0.616	−44.45	−37.65	−34.54	−34.51	−33.71
		Xi34-42	T_3y	53.85	12.92	10.83	2.03	4.93	0.65	9.86	0.637	−49.60	−39.80	−34.5		−33.00
		Zhuang22-21	T_3y	56.21	12.85	11.30	3.42	2.63	0.21	4.18	0.651	−49.40	−38.40	−33.80	−34.00	−33.20

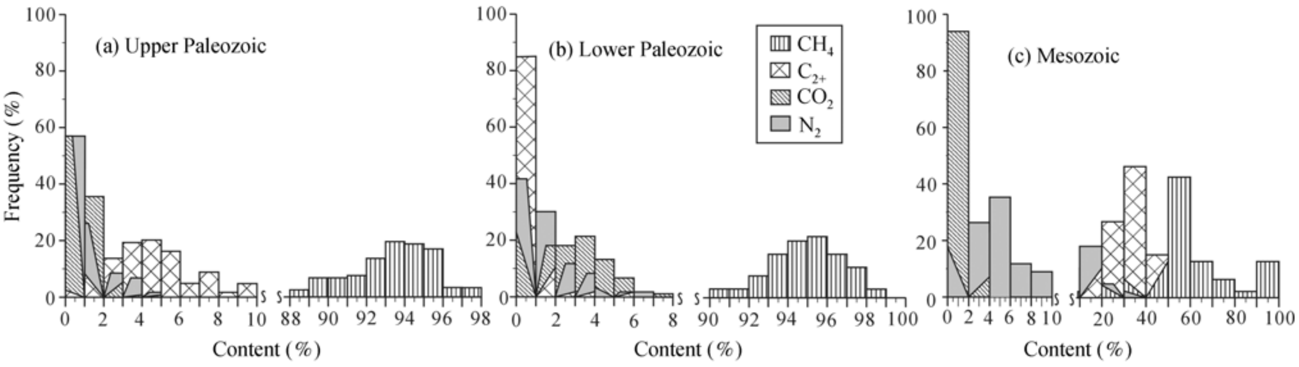

Figure 2 Histograms showing the distribution of gas components: (a) the Upper Paleozoic; (b) the Lower Paleozoic and (c) the Mesozoic.

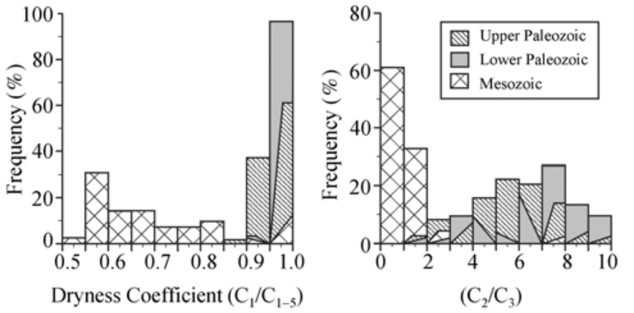

Figure 3 Histograms showing the distribution of the ratios of C_1/C_{1+} and C_2/C_3 of natural gas in the Ordos Basin.

In the Upper Paleozoic, methane is in range of 82.34%—97.91%, with the dominant range of 92%—96% and average of 92.97%; the average concentration of heavy hydrocarbon is 4.61% and the dominant range is 3%—6%; and the dry coefficients (C_1/C_{1-5}) range from 0.847 to 0.992, with average of 0.952, indicating coexistence of dry gas and wet gas.

The methane concentration in the Lower Paleozoic gas is more than that in the Upper Paleozoic. Methane accounts for 90.63%—98.29%, with the dominant range of 93%—98% and average of 94.79%; the average concentration of heavy hydrocarbon is 0.93% and the dominant range is 0—1%; the average value of dry coefficient is 0.990, indicating dry gas.

By contrast, the methane concentration in the Mesozoic is the lowest. Methane concentration ranges from 26.37% to 98.08%, with the dominant range of 50%—60% and average of 59.68%; the average concentration of heavy hydrocarbon is 28.43% and dominant range is 20%—40%; the dry coefficients are in range of 0.380—0.996 with average of 0.671, indicating wet gas.

2.2 Nonhydrocarbon gas contents

The nonhydrocarbon gases in the Ordos Basin are mainly CO_2 and N_2, and their contents are very low. The average values of CO_2 in the Upper, Lower Paleozoic and the Mesozoic are 1.07%, 2.60% and 0.76%, respectively. The average values of N_2 in the Upper and Lower Paleozoic are 1.33% and 1.70%, while the N_2 concentration in the Mesozoic is slightly higher, with an average of 7.38%.

2.3 Ratio characteristics of some components of gaseous alkane (e.g. C_1/C_{1+} and C_2/C_3)

To some extent, the component contents and their ratios can indicate the origin of natural gas. Some useful ratio indexes are listed in Table 2[17]. As shown in Table 1, the $\delta^{13}C_1$ values of gas in the Upper and Lower Paleozoic range dominantly from –40‰ to –30‰, indicating without biogenic gas[18−20].

As shown in Tables 1, 2 and Figures 2, 3, the majority of the ratios of C_2/C_3 in the Upper Paleozoic are over 3, and only few below 3. It could be concluded that the Upper Paleozoic gases are derived mainly from humic cracking gases if Table 2 standards are referred here. In addition, methane concentration in the Upper Paleozoic is mainly over 90%; the ratios of C_1/C_{1+} >0.95 account for near 60%, and the rest of C_1/C_{1+} are between 0.9—0.95, indicating mixing of humic cracking gas and humic pyrolytic gas in the Upper Paleozoic reservoirs.

In the Lower Paleozoic, the majority of methane concentration and dry coefficients are over 95% and 0.95, respectively. The ratio of C_2/C_3 is also more than 3. According to these characteristics, the Lower Paleozoic gases are also mainly humic cracking gas.

The Mesozoic gases are mainly associated gas, with the low methane contents. The dominant ranges for

Table 2 Standard of component parameters [17]

Genetic type of natural gas		Methane concentration (%)	C_1/C_{1-5}	C_2/C_3
Biogenic gas		>95	0.95—1	>2
Sapropelic pyrolytic gas	Oil-associated gas	>50	0.5—0.9	0.9—3.5
	Condensate gas	>60	0.6—0.9	0.9—3.0
Humic pyrolytic gas		>80	0.70—0.95	0.8—3.0
Sapropel type cracking gas		>95	0.95—1	1—3
Humic cracking gas		>95	0.95—1	1.5—7

methane and dry coefficients are 50%—60% and 0.55—0.7, respectively; the ratios of C_2/C_3 are less than 3, indicating the Mesozoic gases would be different from the Upper and Lower Paleozoic gases, and be derived mainly from sapropelic source as oil-type gas.

3 Stable carbon isotopic compositions of natural gas in the Ordos Basin

The chemical compositions of natural gas are usually affected by temperature, pressure, migration, occurrence, biodegradation and so on. When the origin of gas is only determined by chemical composition, the result would be right but sometimes be wrong[21]. The stable carbon isotope has been widely and commonly used for identifying the gas origin.

3.1 Carbon isotopes of gaseous alkane

The carbon isotopes of 192 gas samples (85 gas samples from the Upper Paleozoic reservoirs, 59 gas samples from the Lower Paleozoic and 48 gas samples from the Mesozoic) were collected in this paper (Table 1, Figure 4).

The $\delta^{13}C_1$ values of the Upper Paleozoic gases are −38.47‰ to −29.00‰, with average of −33.45‰. In the Lower Paleozoic, the $\delta^{13}C_1$ values are in range of −41.74‰ to −30.66‰ with average of −34.18‰, which is 0.731‰ less than that in the Upper Paleozoic. As shown in Figure 4(a), the $\delta^{13}C_1$ values of gases from the Upper and Lower Paleozoic reservoirs are enriched in ^{13}C, and the dominant distribution is similar, from −36‰ to −32‰. Contrarily, the $\delta^{13}C_1$ values of the Mesozoic gases are enriched in ^{12}C, ranging from −59.69‰ to −36.67‰, within dominant range of −50‰ to −42‰ and average of −48.70‰. Dai[18] considered the $\delta^{13}C_1$ values of oil-associated gas should be −55‰—−45‰ and the heaviest $\delta^{13}C_1$ value could be −40‰; the $\delta^{13}C_1$ values of condensate oil and gas should be −45‰—−37‰. Thus, the Mesozoic gases are mainly oil-associated gas, mixed with a slight of condensate gas.

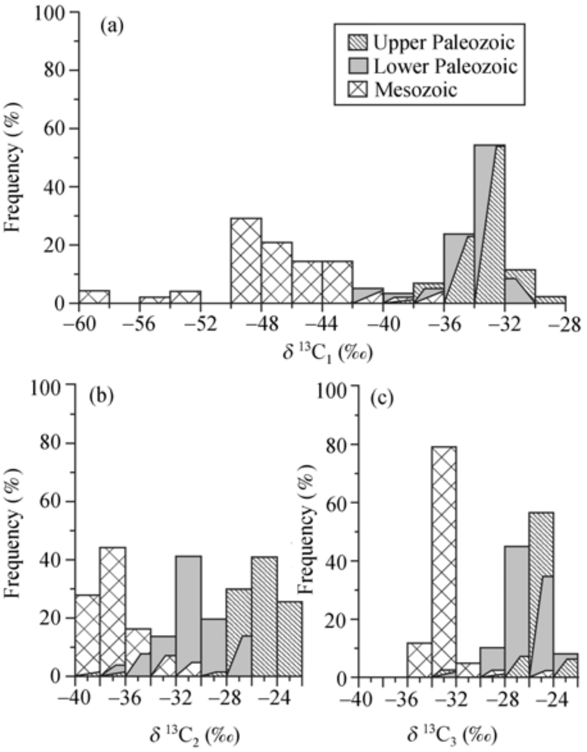

Figure 4 Histograms showing the distribution of $\delta^{13}C_1$, $\delta^{13}C_2$ and $\delta^{13}C_3$ values of the gases from the Upper Paleozoic, Lower Paleozoic and Mesozoic reservoirs in the Ordos Basin.

It is clear from Figure 4(b) that $\delta^{13}C_2$ values of the Upper Paleozoic gases are obviously heavier than those in the Lower Paleozoic. The $\delta^{13}C_2$ values of the Upper Paleozoic gases dominantly range from −28‰ to −22‰ with average of −25.20‰, while $\delta^{13}C_2$ values in the Lower Paleozoic dominantly range from −34‰ to −26‰ with average of −30.09‰. The $\delta^{13}C_2$ values of the Mesozoic gases are less than those in the Upper and Lower Paleozoic, with the dominant range of −40‰ to −34‰ and average of −36.70‰. Many scientists have defined the standards of $\delta^{13}C_2$ values between coal-formed gas and oil-type gas. For example, Zhang,

Zhang[22] and Dai[19,23] proposed that the standard of $\delta^{13}C_2$ value is –28‰, –29‰ and –28.8‰, respectively. Accordingly, the gases in the Upper Paleozoic and Mesozoic reservoirs are typically coal-formed gases and oil-type gases, respectively. But natural gas with isotopically lighter ethane and heavier methane in the Lower Paleozoic can hardly determine the gas origin.

As shown in Figure 4(c), the $\delta^{13}C_3$ values of the Upper Paleozoic gases are a little heavier than those of the Lower Paleozoic, and the $\delta^{13}C_3$ values of the Mesozoic gases are the lightest among the three strata. The dominant ranges of $\delta^{13}C_3$ values of gases from the Upper, Lower Paleozoic and Mesozoic reservoirs are –26‰ — –24‰, –28‰— –24‰ and –34‰——32‰, respectively, and their average values are –24.42‰, –25.75‰ and –32.94‰, respectively. Zhang (1986) proposed that $\delta^{13}C_3$ values of coal-formed gas would range from –26‰ to –18‰ and those of oil-type gas from –32‰ to –28‰. Dai[19,23] defined that $\delta^{13}C_3$ values of oil-type gas would be less than –25.5‰. Thus, it is clear from the $\delta^{13}C_3$ values that gases from the Upper Paleozoic and the Mesozoic are mainly coal-formed gas and oil-type gas, respectively, consistent with the result based on the standards of $\delta^{13}C_1$ and $\delta^{13}C_2$ values. But, the type of the Lower Paleozoic gas cannot be determined because the $\delta^{13}C_3$ values are distributed between the standards of coal-formed gas and oil-type gas.

3.2 Difference of the $\delta^{13}C$ values of gases from different gas fields in the Upper Paleozoic

Based on the elevated $\delta^{13}C$ values, the Upper Paleozoic gases are dominated by coal-formed gas. But it should be noted that there is a little difference of the $\delta^{13}C$ values among different gas fields in the Upper Paleozoic. As shown in Figure 5, among three main gas fields in the Upper Paleozoic, the $\delta^{13}C_2$ values of gases from the Yulin gas field are the lightest. It could be well explained that a few oil-type gases produced from the C-P carbonates would be mixed in the Yulin gas field, because the C-P carbonates are mainly distributed in the middle and east of the Basin and the Yulin gas field is just located in the east of the Basin.

3.3 Discussion on the $\delta^{13}C_2$ and $\delta^{13}C_3$ values of the Lower Paleozoic gas

As shown in Figure 6, the carbon isotope compositions of gases from the Upper Paleozoic, Lower Paleozoic and the Mesozoic are different in the Ordos Basin, suggesting the different types of natural gases. Based on the above, the Mesozoic gases are of oil-type gas, different from the gases in the Upper and Lower Paleozoic reservoirs, but it is hard to well differentiate the genesis types between the Upper and Lower Paleozoic gas.

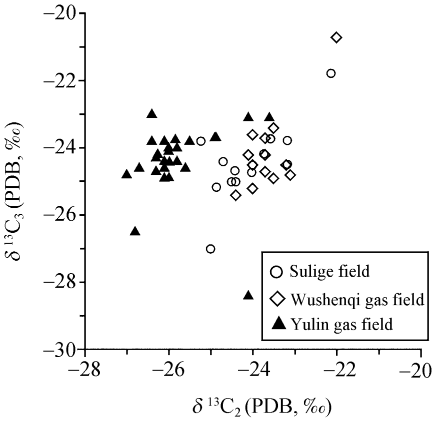

Figure 5 Chart showing the distribution of $\delta^{13}C_2$ and $\delta^{13}C_3$ values of gases from different gas fields in the Upper Paleozoic.

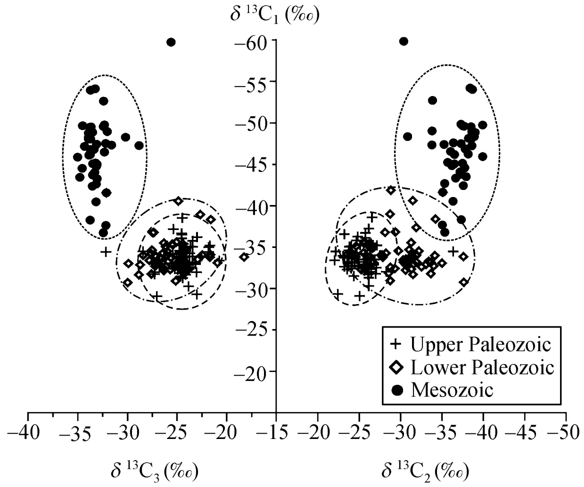

Figure 6 Chart showing the distribution of $\delta^{13}C_1$- $\delta^{13}C_2$- $\delta^{13}C_3$ of the gases from the Upper Paleozoic, the Lower Paleozoic and the Mesozoic reservoirs in the Ordos Basin.

As shown in Figure 6, there are two main differences of the $\delta^{13}C_n$ values between the Upper and Lower Paleozoic gas: ①The ranges of $\delta^{13}C_n$ values of the Lower Paleozoic gases are wider than those of the Upper Paleozoic, indicating that gas source would be single one in the Upper Paleozoic but complex in the Lower Paleozoic[24]; ② The $\delta^{13}C_2$ and $\delta^{13}C_3$ values of the Lower Paleozoic gases are much less than those in the Upper Paleozoic. If $\delta^{13}C_2$ = –29‰ is considered as standard to differentiate coal-formed and oil-type gas, the Upper

Paleozoic gases are considered as coal-formed gas with $\delta^{13}C_2>-29$‰; but the Lower Paleozoic gases with both $\delta^{13}C_2>-29$‰ and $\delta^{13}C_2<-29$‰ are observed, thus, it is hard to determine which type is dominant in the Lower Paleozoic. Some scientists considered that the Lower Paleozoic gases with $\delta^{13}C_2<-29$‰ are of oil-type gases[11], but that using $\delta^{13}C_n$ values to determine gas origin is only available for single source gas and not available for multiple source gas. The complex carbon isotopic characteristics of the Lower Paleozoic gases suggest that the gases would be from multiple sources. In combination with the previous achievement, the main question will be discussed as follows.

Firstly, when the mixed gases are dominated by coal-formed gas with a slight of sapropelic gases, the $\delta^{13}C_2$ values would become less than -29‰. This opinion has been discussed, for example, Xia[25] calculated the changes of $\delta^{13}C_2$ values when the gases mixed with different proportions, and proposed that $\delta^{13}C_2$ values would show the characteristic of oil-type gas when the mixing ratio of coal-formed gas and oil-type gas is less than 8. Dai[11,24] considered that coal-formed gas contained more methane and less ethane than oil-type gas, so when they are mixed, the concentration of oil-type ethane would be more than that of coal-formed ethane and the $\delta^{13}C_2$ values of mixture would be similar to the oil-type gas.

Secondly, fractionation including component fractionation and isotope fractionation during migration would lead to the change of carbon isotope[26−30]. Xia[25] proved its existence that fractionation of mixed gases (mixing of coal-formed gas and oil-type gas) would lead $\delta^{13}C_2$ values to be lighter and even show the characteristics of oil-type gas.

Based on the above explanation, if the gases in the Lower Paleozoic were mixed gases dominated by coal-formed gases with a slight of oil-type gases and mainly derived from the source rocks of the Upper Paleozoic, it would be well understood why $\delta^{13}C_1$ values of the Lower Paleozoic gases are similar with those of the Upper Paleozoic but $\delta^{13}C_2$ values are much lighter.

That $\delta^{13}C_3$ values in the Lower Paleozoic less than those in the Upper Paleozoic is also explained as similar law above. But, the difference of $\delta^{13}C_3$ values between the Upper and Lower Paleozoic is less than that of $\delta^{13}C_2$ values ($\delta^{13}C_{2\mathrm{Upper}}-\delta^{13}C_{2\mathrm{Lower}}=4.783$‰, but $\delta^{13}C_{3\mathrm{Upper}}-\delta^{13}C_{3\mathrm{Lower}}=1.241$‰), because the difference of $\delta^{13}C_n$ values between coal-formed gas and oil-type gas from different types of kerogen with similar maturity would become less with the increase of carbon number[23].

3.4 Discussion on the source rocks maturity and gas origin

The $\delta^{13}C_1$-R_o (vitrinite reflectance) relationship could be used to distinguish the humic and sapropelic methane, and their regression equations are as follows: $\delta^{13}C_1\approx 14.121\ \mathrm{g}R_o-34.39$ (humic); $\delta^{13}C_1\approx 15.801\ \mathrm{g}R_o-42.20$ (sapropelic)[23,31]. Statistically, $\delta^{13}C_1$ values ranging from -35.55‰ to -30.66‰ in the Lower Paleozoic account for 86.44%. As shown in Figure 7, if these 86.44% gas samples are of oil-type gases, it could be concluded that the R_o values should be within range of A″—B″, more than 2.65%, and most more than 3%. But the previous results indicated that R_o values of the Lower Paleozoic source rocks were in range of 2.07%—2.68%, whereas the R_o values of the Upper Paleozoic source rocks are less. Thus, the oil-type gas should not be the main type in the Lower Paleozoic. When methane generated from humic source is assumed as a main component, the R_o values of gases with $\delta^{13}C_1$ values between -35.55‰—-30.66‰ are in range of 0.83%—1.84%, almost consistent with the R_o values of the Carboniferous-Permian coal. Thus, methane in the Lower Paleozoic should be derived mainly from C-P coal measures. The $\delta^{13}C_1$ values of the rest gas samples in the Lower Paleozoic are in range of -41.74‰—-36.67‰. As shown in Figure 7, the R_o values of the rest gases calculated from the regression equation of oil-type methane are in range of 1.1%—2.2%, similar

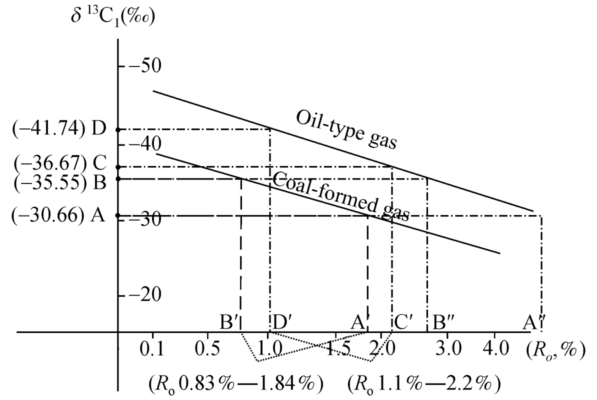

Figure 7 $\delta^{13}C_1$–R_o chart discriminating humic methane and sapropelic methane.

with the thermal maturity of the Upper Paleozoic source rocks, too. Accordingly, the methane in the Lower Paleozoic should be derived dominantly from C-P coal, mixed with a slight of oil-type methane from the Upper Paleozoic sapropelic sources.

As shown in Figure 4, $\delta^{13}C_1$ values of the Upper Paleozoic gases are a little heavier than those of the Lower Paleozoic gases, so humic methane accounts for the most part in the Upper Paleozoic which are of self-generation and self-reservation.

In the Mesozoic, the majority of $\delta^{13}C_1$ values are less than −40‰. Based on the $\delta^{13}C_1$-R_o relationship, it is easily concluded that the gases are of oil-type gases.

3.5 Stable carbon isotope reversal of gaseous alkane

The carbon isotopic values of C_1 to C_4 alkanes can be in a normal order of $\delta^{13}C_1<\delta^{13}C_2<\delta^{13}C_3<\delta^{13}C_4$ or in a reversal order (i.e, $\delta^{13}C_1>\delta^{13}C_2>\delta^{13}C_3>\delta^{13}C_4$). If a reversal occurs only between two immediate homologues, it is called a singular reversal. Likewise, a multiple reversal is usually referred if the reversal involves more than two homologues. While the cause for singular reversal is simple, the situation for multiple reversal is more complicated[3]. Dai[32,33] proposed four possible reasons for stable carbon isotope reversal of gaseous alkanes, involving the mixing of biogenic and abiogenic gas, mixing of sapropelic and humic gas, mixing of gases from two source rock intervals of similar kerogen type but of different maturity (or from one source rock unit of varying maturity), or microbial oxidation.

As shown in Figure 8(a), gases from the Sulige and Wushenqi fields display a singular reversal of $\delta^{13}C_2>\delta^{13}C_3$, whereas gases in the Yulin field exhibit singular reversal of $\delta^{13}C_3>\delta^{13}C_4$. A significant contribution from oil-type gas is excluded in the Sulige, Wushenqi and Yulin fields, as they are produced mainly from coal measures in the Lower Permian Shanxi Formation. As the Ordos Basin is one of the most tectonically stable basins in China and has experienced little faulting, the mixing of biogenic and abiogenic gas is incomprehensible. The component reversal of two immediate homologues which exhibit carbon isotopes reversal does not happen in these gas fields, so microbial oxidation is unlikely, either[34]. Therefore, the most likely cause for the singular isotope reversal of the Upper Paleozoic gases was the mixing of gases from humic sources with different thermal maturity levels. Besides, gases in the Upper Paleozoic display $\delta^{13}iC_4>\delta^{13}nC_4$.

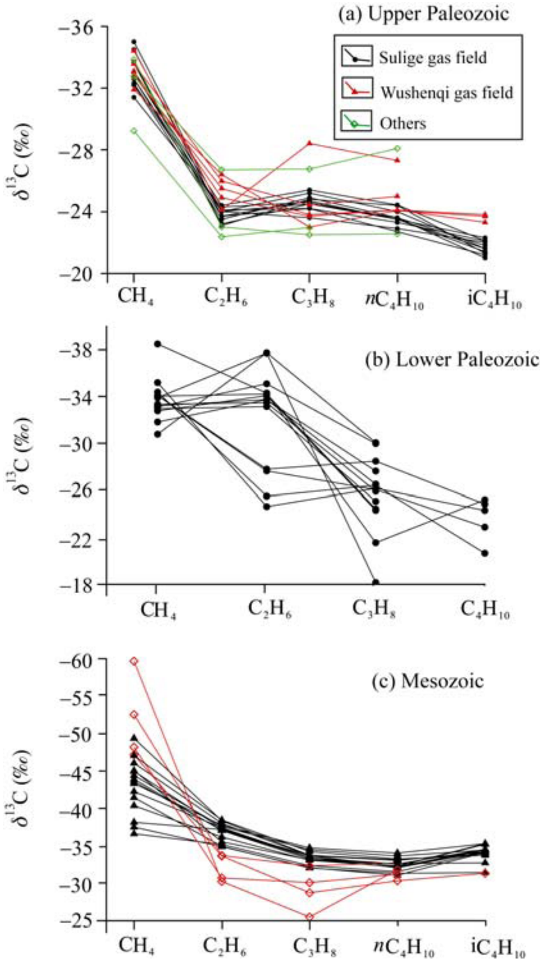

Figure 8 Broken line graph of the carbon isotope of methane and its homologues of gases in the Ordos Basin.

As shown in Figure 8(b), the Lower Paleozoic gases exhibit the multiple reversals in carbon isotopes (i.e. $\delta^{13}C_1>\delta^{13}C_2$, $\delta^{13}C_2>\delta^{13}C_3$ and $\delta^{13}C_3>\delta^{13}C_4$), and the reversal ranges in the Lower Paleozoic are wider than those in the Upper Paleozoic. The isotope reversal for propane and butane (i.e. $\delta^{13}C_3>\delta^{13}C_4$) is commonly observed, the reversal for ethane and propane (i.e. $\delta^{13}C_2>\delta^{13}C_3$) is few and for methane and ethane (i.e. $\delta^{13}C_1>\delta^{13}C_2$) is very rare[35]. Fuxe[35] proposed that one possible reason for this rare reversal for methane and ethane is the increase in the proportion of gases with high maturity formed during the later evolution stage. But we propose that the possible reason for the reversal of methane and ethane here should be mixing of high maturity sapropelic and humic gas.

The stable carbon isotopes of methane to propane in the Mesozoic is in a normal order of $\delta^{13}C_1<\delta^{13}C_2<\delta^{13}C_3$, and only few samples show the single reversals of $\delta^{13}C_3>\delta^{13}C_4$ (Figure 8(c)). Some isotope reversals of $\delta^{13}C_3>\delta^{13}C_4$ are caused by the microbial oxidation because the gas displays components reversal of two homologues which exhibit carbon isotopes reversal. For example, the carbon isotope values of propane and butane from Well Hu 401 are −30.16‰ and −31.26‰, and the corresponding component contents are 0.6% and 2.71%, respectively (Table 1). The similar observation occurs in Well Ma 9-1. The other isotope reversals of $\delta^{13}C_3>\delta^{13}C_4$ may be caused by mixing of gases from sapropelic sources of different maturity. Besides, in contrast to the carbon isotopic characteristics of butane in the Upper Paleozoic, there is a reversal of $\delta^{13}iC_4<\delta^{13}nC_4$ observed from the Mesozoic (Figure 8(c)). The authors considered the different isotopic characteristics for *n*-butane and *iso*-butane between the Upper Paleozoic and the Mesozoic may be caused by different types of kerogen. The gases in the Upper Paleozoic were mainly produced from humic type kerogen, whereas the gases in the Mesozoic were from sapropelic type kerogen.

4 Gas-source correlations in the Ordos Basin

Based on the chemical and carbon isotopic compositions of gaseous alkanes, natural gases in the Upper Paleozoic were derived mainly from humic sources, such as Carboniferous-Permian coal measures. There is also a slight contribution of oil-type gases derived from the Upper Paleozoic carbonates in Yulin gas field. The Mesozoic gases are of mainly oil-associated gas, mixed with a little condensate gas. The sapropelic source in the Triassic Yanchang Formation is a main source.

However, there are several opinions about the source of the Lower Paleozoic gases. Yang[2] proposed that the Lower Paleozoic gases were the mixing of coal-formed gases produced from the Upper Paleozoic source rocks and oil-type gases derived from the Lower Paleozoic sources, but did not point out which source was a main contributor. Guan[6] suggested that the gases in the Upper and Lower Paleozoic reservoirs were derived from the same source due to the similar distribution of $\delta^{13}C_1$ values. Zhang[7] considered that the Ordovician gases were migrated from the Upper Paleozoic, but Huang[8] and Chen [9] considered that the carbonates in the Lower Ordovician Majiagou Formation were the dominant source for the Lower Paleozoic gases based on the $\delta^{13}C_2$ values. Cai[36] discussed the geochemical characteristics of gas together with the effect of TSR, and pointed out that the Ordovician gases were mixed gases produced from the C-P coal measures and Ordovician carbonates. Dai[11] proposed that the Lower Paleozoic gases were mixing of majority coal-formed gases produced from C-P coal measures and minority oil-type gases from the Upper Paleozoic carbonates.

Based on the above discussion, it could be concluded that the Lower Paleozoic gases are dominated by coal-formed gases produced from C-P coal, mixed with a slight of oil-type gases. Two aspects about the source of oil-type gases in the Lower Paleozoic will be discussed here. Firstly, based on the $\delta^{13}C_1$-R_o relationship, the thermal maturity of the source rocks which produced oil-type gases in the Lower Paleozoic did not reach the maturity level of the Ordovician carbonates, but was similar with the Upper Paleozoic source rocks. Secondly, the organic-lean carbonates with an average of 0.22% TOC in the Lower Paleozoic could not be the effective gas source. Xia[37] considered that the Ordovician carbonates in Ordos Basin were a poor source; Liang[38] analyzed hydrocarbon potential of source in the Tarim Basin, and proposed that the economic oil and gas discoveries with a carbonate source occur mostly adjacent to organic-rich source rocks (with TOC>0.5%); Zhang[39] pointed out that no economic oil or gas field has been encountered in the regions where the only possible source rocks are carbonates with TOC<0.2%; Wang[40] also proposed that carbonate with TOC⩾0.5% was a available source; Tissot[41] and Bjorlykke[42] suggested that the TOC for carbonates as an active source for economic gas and oil should be more than 0.3% and 0.5%, respectively. Thus, the carbonates in the Lower Ordovician Majiagou Formation are unlikely a significant contributor to the oil-type gases in the Lower Paleozoic, whereas the carbonates in the Upper Paleozoic might be a main gas source with TOC of 0.5%－3%.

5 Conclusions

The focus of the current investigation was the origin of the gases in the Upper Paleozoic, Lower Paleozoic and Mesozoic in the Ordos Basin. The chemical and stable carbon isotopic compositions of gas samples from these strata were discussed, and then the conclusions are drawn as follows:

(1) In the Upper Paleozoic, the methane is a main component, accounting for more than 90%; the ratios of

C_2H_6/C_3H_8 in most samples are more than 3; the $\delta^{13}C_n$ values are enriched in ^{13}C, indicating the gases are mainly coal-formed gases. The singular reversal in the carbon isotopes of gaseous alkanes happen widely due to the mixing of gases from III kerogen with different thermal maturity levels. Besides, $\delta^{13}C_2$ values of gases from the Yulin gas field are a little lighter than those in the other gas fields because there might be a little mixing of oil-type gases in the Yulin field.

(2) In the Lower Paleozoic, the methane in most gas samples account for more than 95%; the ratios of C_2/C_3 are more than 3; the $\delta^{13}C_1$ values are heavy but $\delta^{13}C_2$ and $\delta^{13}C_3$ values are much lighter, suggesting that the Lower Paleozoic gases would be dominated by coal-formed gases mixed with a slight of oil-type gases. In the Upper Paleozoic, there is multiple reversal of carbon isotopes especially very rare reversal for methane and ethane (i.e. $\delta^{13}C_1>\delta^{13}C_2$), which may be caused by the mixed gases from the high maturity sapropelic and humic sources.

(3) In the Mesozoic, the dominant range for methane is 50%—60%; the ratios of C_2/C_3 are less than 3; the $\delta^{13}C_n$ values are obviously enriched in ^{12}C, indicating the gases would be of oil-associated gases. The carbon isotopic reversal for propane and butane is caused by microbial oxidation and mixing of gases from sapropelic source of different maturity. In comparison with the gases in the Upper Paleozoic, the Mesozoic gases with $\delta^{13}iC_4>\delta^{13}nC_4$, may be caused by different types of kerogen.

(4) Based on the $\delta^{13}C_1$-R_o relationship and extremely low total organic carbon contents, it could be concluded that the gas accumulations in the Lower Paleozoic were produced mainly from Upper Paleozoic source rocks, not from the Lower Paleozoic.

We would like to thank Professor Dai Jinxing for his advice and supervising

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