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Research Article

Geochemical Characteristics of Natural Gas and Hydrocarbon Charge History in the Western Qaidam Basin, Northwest China

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31 natural gases in the western Qaidam Basin of China were collected and analyzed for gas composition including light hydrocarbons (C_5-C_7) and carbon isotopic characteristics. Based on genetic type obtained from C_1-C_3 and C_7 fractions, four types of gases are identified: oil-type gas, coal-type gas, biodegraded gas, and mixed gas. The oil-type gas is the predominant-type gas in the western Qaidam Basin; coal-type gas is mainly distributed in the Zhahaquan and Nanyishan fields; mixed gas is mainly in the Zhahaquan, Wunan, and Nanyishan fields; and biodegraded gas is mainly distributed in the Huatugou and Yuejinerhao fields. According to the empirical relationship between $\delta^{13}C_1$ and the equivalent vitrinite reflectance (R_o, %) of source rock, the R_o values of gas range from 0.6% to 1.5%, with an average value of 0.9%. The generation temperatures of major reservoired hydrocarbons (GTMRH) calculated from the C_7 components range from 115.6°C to 141.7°C, with an average value of 126.5°C. These two maturity indicators have relatively positive correlation and reveal that the maturity of gas increases from west to east in the southwestern Qaidam Basin. Moreover, combining GTMRH with the homogenous temperature of petroleum inclusions, it is inferred that major petroleum charge in the western Qaidam Basin mainly occurred during the late period of the Himalayan movement. Deep hydrocarbon fluid sources were found in the Shizigou, Yingdong, Zhahaquan, and Nanyishan fields; thus, the deep reservoirs of paleouplifts adjacent to the hydrocarbon-generating depressions are estimated as a favorable area for further exploration in the western Qaidam Basin.

1. Introduction

The Qaidam Basin is the largest petroliferous sedimentary basin inside the Tibetan Plateau, northwest China, and significant amounts of oil have been produced in the western Qaidam Basin [1]. In recent years, oil and gas exploration has made considerable progress in the Yingdong, Zhahaquan, and Yingxi fields in the western Qaidam Basin. Some newly produced oil has lower viscosity and density with a higher gas-oil ratio, suggesting that they may be derived from deeper strata. However, little research has been conducted to investigate the genetic type and maturity of these newly produced gases. Furthermore, a systematic analysis of natural gas is necessary to deepen the recognition of gas origin and charge history in the western Qaidam Basin.

The hydrocarbon components of natural gas mainly consist of C_1-C_5 compounds with trace C_{5+} light hydrocarbon compounds. The definition of light hydrocarbon has not been unified. In early studies, light hydrocarbons refer to hydrocarbons ranging from C_1 to C_{14} [2–4]; in later studies, the ones refer to hydrocarbons ranging from C_5 to C_{10} [5–7]. The C_5-C_7 light hydrocarbons are the focus in current researches and this paper. Besides, partial light hydrocarbons would dissolve in the gas phase because natural gas can be taken as the solvent and the volatility of light hydrocarbon. Thus, light hydrocarbons could distribute in the gas phase and liquid phase in a gas sample. Previous studies reported that separation of the oil phase and the gas phase mainly affects the aromatic content of a light hydrocarbon, with little effect on most light hydrocarbon parameters [2, 8, 9]. So far, light hydrocarbons in dry gas and wet gas have been successfully analyzed [6, 7]. Many studies have applied the light hydrocarbon as an ancillary geochemical tool to evaluate the genetic type and thermal maturity of natural gas and identify secondary alteration [3–7]. Furthermore, natural gas in western Qaidam Basin is wet gas with a higher content of light hydrocarbons, which could better reflect the geochemical characteristics of natural gas.

Identification of the genetic type of natural gas is important for assessment of its sources and exploration potential [10]. Based on sources, there are two broad categories: biogenic and abiogenic gases. Abiogenic gas includes gas from the mantle and abiogenic formation. Biogenic gas includes biogenic gas and thermogenic gas, and the latter can be further divided into coal-type gas and oil-type gas based on the type of organic matter [11, 12]. Many researchers have already established some geochemical parameters and classic diagrams to identify the genetic type of natural gas [12–16]. The composition of natural gas and methane carbon isotope values ($C_1/(C_2+C_3)$ vs. $\delta^{13}C_1$) is significant in identifying different types of natural gas [13]. Besides, the carbon isotope of ethane is widely used to distinguish the coal-type gas and the oil-type gas [14], including the cross plot of $\delta^{13}C_1$-$\delta^{13}C_2$-$\delta^{13}C_3$, $\delta^{13}C_1$-$\delta^{13}C_{CO2}$, $\delta^{13}C_{2-3}$-$\ln(C_2/C_3)$, and so on [12, 15, 16]. Natural gas in the western Qaidam Basin is mainly derived from saline lacustrine depositions which are enriched in $\delta^{13}C_2$, and Zhang et al. [17] modified the classification criteria for coal-type gas and oil-type gas. Owing to the complex formation process of natural gas and secondary alteration, the genetic identification of different types of natural gas should be based on multiple parameters. C_5-C_7 light hydrocarbons can be used to provide some new information insight into natural gas generation. Therefore, a detailed study of natural gas could get a better understanding of its genetic type.

Thermal maturity has an important effect on the composition and carbon isotope of natural gas. The carbon isotope values of methane increase with increasing thermal maturity of source rocks and are adapted to study the maturity of gas [18, 19]. Besides, the dryness coefficient of natural gas increases with an increase of thermal maturity. Moreover, light hydrocarbons could be used to calculate the expulsion temperature of oil and gas [20]. The ratio of 2,4-dimethylpentane to 2,3-dimethylpentane (2,4-DMC_5/2,3-DMC_5) was found related to temperature, and it was calibrated to expulsion temperature [8, 21]. Studies have reported that expulsion temperatures have positive correlations with biomarker maturity parameters and the gas-oil ratio [22–24]. While little research has been conducted to evaluate the expulsion temperatures of natural gas in the western Qaidam Basin, it may provide a novel way of assessing the maturity of gas.

The homogenous temperatures of petroleum inclusions are always applied to reconstruct the petroleum charging history combining with burial history and hydrocarbon generation history. Numerous researches have been carried out, and most suggest that there are primarily two petroleum charging episodes in the western Qaidam Basin [25–30]. Because each hydrocarbon charge event has distinct contributions to reservoired petroleum, it is important to recognize that a major charge event is not only of academic significance but also related directly to the evaluation of the commercial potential in a studied area. Dieckmann et al. [20] proposed that expulsion temperature calculated from the C_7 light hydrocarbon could reflect the average temperature of hydrocarbon expulsion. Generally, the reservoired petroleum is composed of hydrocarbons from multiperiod charging and dominated by the ones from the major period, so that the expulsion temperature obtained from the C_7 light hydrocarbon, which is collected from reservoired petroleum, could be considered as the generation temperature of major reservoired hydrocarbons (GTMRH). The GTMRH reflects the character of reservoired petroleum and may provide information about major petroleum charging.

In this study, 31 natural gases in the western Qaidam Basin of China were collected and analyzed for gas composition and carbon isotopic characteristics. The aims of this study are (1) to determine the genetic type and thermal maturity of natural gas in the western Qaidam Basin, (2) to investigate the application of GTMRH to petroleum charging history, and (3) to provide suggestions for future hydrocarbon exploration.

2. Geological Setting

The Qaidam Basin is located in the northeastern Tibetan Plateau of northwest China and covers a total area of 121,000 km^2 (Figure 1(a)). It contains Mesozoic and Cenozoic lacustrine sedimentary sequences that were deposited on the Qaidam block during the pre-Jurassic period, and maximum sediment thickness could reach 17,280 m [1]. The basin is bounded by the Qilian Mountains to the northeast, the Altun Mountains to the northwest, and the Kunlun Mountains to the south. The basin can be structurally divided into three tectonic units, namely, the Northern Fault-Block Belt, the Western Depression, and the Eastern Depression. This study was carried out in the Western Depression (Figure 1(b)). Besides, the western Qaidam Basin includes two parts: the northwestern and the southwestern Qaidam Basin, and gas samples in both parts are obtained in this study (Figure 1(c)).

In the western Qaidam Basin, the Tertiary saline lacustrine depositions are primary hydrocarbon source rocks. The source rocks are lacustrine mudstone and marlstone in the Lower Xiaganchaigou-Shangganchaigou Formations. The main reservoirs are sandstone in the Lulehe-Shangyoushashan Formations and marlstone in the Upper Xiaganchaigou Formation. The cap rocks are mudstone in the Lulehe-Shizigou Formations and marlstone in the Upper Xiaganchaigou Formation (Figure 2).

Source rocks in the western Qaidam Basin have relatively low amount of total organic carbon (TOC), 0.1-2.7% (average below 1.0%), but their generation potentials reach a fairly high level with chloroform extraction mainly between 0.05% and 0.5% [31]. The organic matter of two intervals is

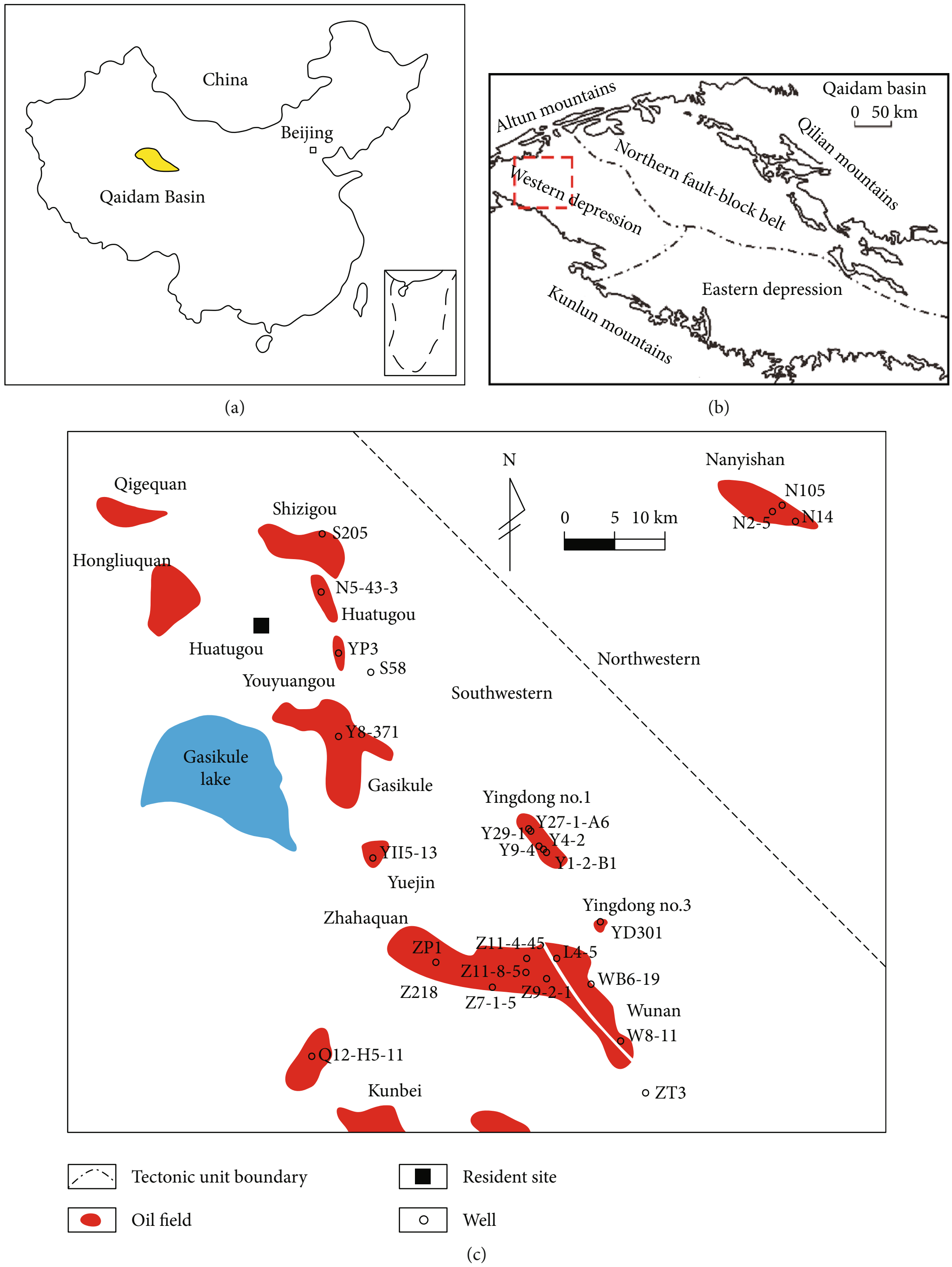

FIGURE 1: (a) Location of the Qaidam Basin within China; (b) location of three tectonic units in the Qaidam Basin and the rectangle indicates the studied area; (c) distribution of oil fields and sample sites and the dashed line is the boundary between southwestern and northwestern Qaidam Basin.

mainly type-II kerogen and a little of type-III kerogen [32]. The vitrinite reflectance values (R_o, %) of these source rocks range from 0.4% to 1.3% [31]; corresponding oils within the immature to mature stages are all discovered in the western Qaidam Basin [33, 34].

3. Samples and Methods

3.1. Sampling. Natural gases were sampled in the major oil fields, e.g., Shizigou, Huatugou, Youyuangou, Gasikule, Yuejinerhao, Yingdong (including no. 1 and no. 3), Wunan,

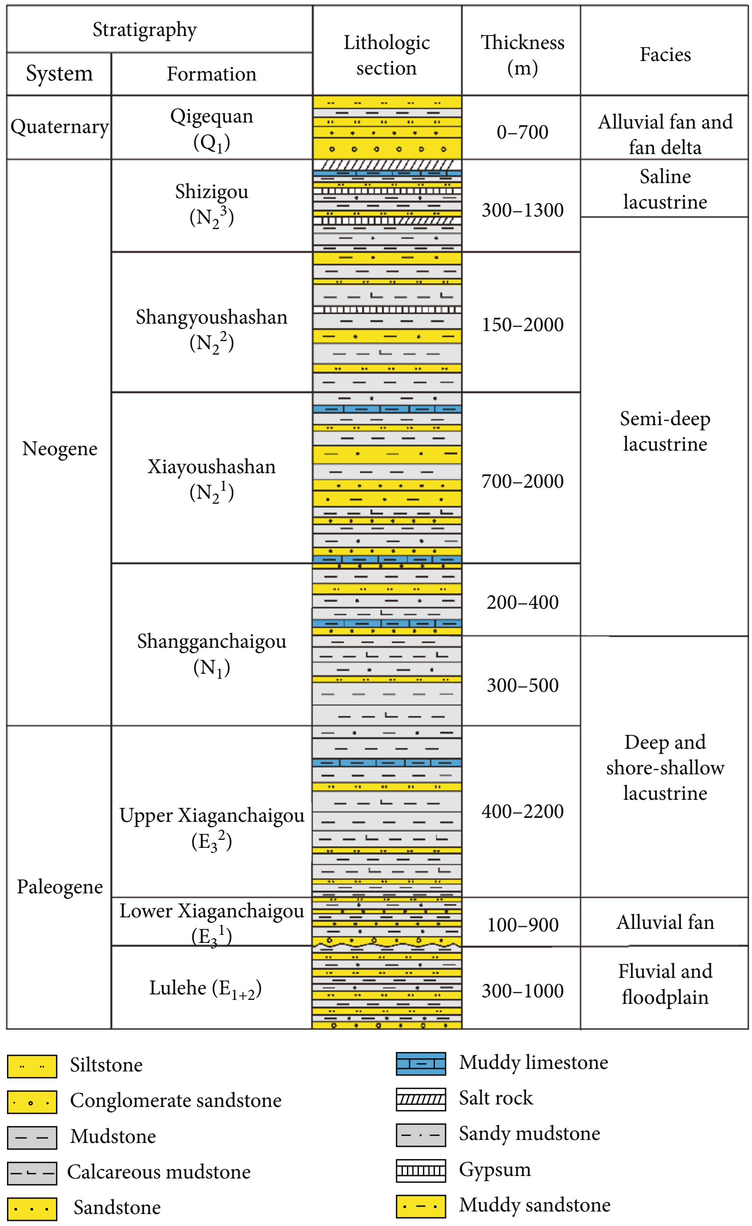

Figure 2: Generalized stratigraphic column for the western Qaidam Basin (modified after [46]).

Zhahaquan, Kunbei, and Nanyishan oil fields (Figure 1(c)). Gas samples were directly collected at wellheads in the commercial oil production fields using aluminum alloy cylinders with double valves. Before sampling, cylinders were flushed three times at the wellhead to remove air. In total, 31 natural gases were sampled in the western Qaidam Basin, and the sample data are listed in Table 1.

Because of the volatility of light hydrocarbons, evaporation is inevitable during sampling, storage, and sample preparation when using crude oil to analyze light hydrocarbon composition, and even minor evaporation will affect light hydrocarbon parameters [35], while natural gas stored in aluminum alloy cylinders has no evaporation, which is essential for the original light hydrocarbon component.

3.2. Gas Composition and Stable Carbon Isotopic Analysis. Gas compositions were determined on a GC-9160 gas chromatograph equipped with two flame ionization detectors and a thermal conductivity detector. Individual hydrocarbon gas components from C_1 to C_5 were separated using a capillary column (PLOT Al_2O_3 $50\,m \times 0.53\,mm \times 0.32\,\mu m$). The GC oven temperature was initially held at 35°C for 5 min and then increased to 200°C at 10°C/min and held at this temperature for 10 min. Gaseous nonhydrocarbons (N_2, CO_2, and H_2S) were analyzed on a high-resolution mass spectrometer with an electric impact ion source, using a selected ion-monitoring method to determine molar percentages, which were converted to molar concentration using the ideal gas function. The emission current was 40 μA, and

Table 1: Representative light hydrocarbon geochemical parameters in natural gas from the western Qaidam Basin.

Field	Well	Depth (m)	Strata	n-C_7/ MCC_6	Toluene/ n-C_7	$\Sigma DMCC_5$ (%)	n-C_7 (%)	MCC_6 (%)	2-MC_6/ 3-MC_6	GTMRH (°C)
Shizigou	S205	3378-3598	E_3^2	1.69	0.40	18.0	51.5	30.5	0.74	115.6
	S58	5451.18	E_3^2	1.23	0.74	20.2	44.0	35.7	0.72	119.5
Huatugou	HN5-43-3	1015	N_1	1.73	0.10	30.4	44.1	25.5	0.83	121.8
Youyuangou	YP3	782.0-783.7	N_2^1	2.72	0.19	13.9	63.0	23.1	1.12	128.5
Gasikule	Y8-371	3481.8-3528.0	E_3^2	2.41	0.23	15.9	59.5	24.7	0.86	124.3
Yuejinerhao	YII5-13	—	E_3^1	1.13	0.49	40.3	31.7	28.0	0.70	126.4
Yingdong no. 1	Y1-2-B1	966-996.1	N_2^2	1.78	0.58	19.3	51.7	29.1	1.18	126.9
	Y1-2-B4	1288.3-1318.1	N_2^2	2.66	0.41	17.8	59.7	22.5	1.12	128.4
	Y27-1-A6	1739-1802	N_2^2	2.81	0.36	15.7	62.2	22.2	1.41	130.4
	Y38-3-C	1271.5-1342.9	N_2^2	2.60	0.53	20.5	57.4	22.1	1.19	127.3
	Y29-1	2838.2-2896.8	N_2^1	2.16	0.62	20.7	54.1	25.1	1.00	124.4
	Y4-2	3018.7-3103.7	N_2^1	2.32	0.36	16.9	58.0	25.0	1.02	127.4
	Y9-4	2081.3-2219.9	N_2^1	2.68	0.39	21.0	57.5	21.5	1.38	132.1
Yingdong no. 3	YD301	2012.2-2023.3	N_2^1	2.14	0.56	20.5	54.2	25.3	1.08	124.2
	YD3-3	2313.3-2491.4	N_2^1	2.22	0.50	20.7	54.6	24.7	1.16	133.3
Wunan	L4-5	2041.6-2305.5	N_2^1	1.89	2.65	16.3	54.7	28.9	1.09	121.4
	W8-11	1361.7-1420.7	N_2^1	1.60	0.77	10.1	55.3	34.5	1.01	130.2
	WB6-19	1631.8-1734	N_2^1	1.37	0.49	18.0	47.4	34.6	0.81	121.3
Zhahaquan	Z11-33-5	—	N_2^1	2.61	0.22	20.1	57.8	22.2	1.11	125.6
	Z11-4-45	—	N_2^1	2.51	0.20	20.6	56.7	22.6	1.11	125.7
	Z11-8-5	2493.0-2494.1	N_2^1	2.04	0.57	19.7	53.9	26.4	1.00	127.2
	Z218	—	N_2^1	2.71	0.39	46.9	38.8	14.3	0.92	129.3
	Z7-1-5	3530.8-3534.7	N_1	2.28	0.29	14.7	59.3	26.0	1.00	129.1
	Z7-2-3	—	N_1	2.28	0.39	15.1	59.0	25.8	1.11	130.1
	Z9-2-1	2432.2-2840.6	N_2^1	1.85	0.51	19.6	52.2	28.2	1.00	125.1
	ZP1	—	N_2^1	1.93	0.37	22.6	51.0	26.5	0.92	128.3
	ZT3	3144-3150	E_3^1	8.77	0.29	5.7	84.6	9.6	1.10	141.7
Kunbei	Q12-H5-11	—	E_3^1	1.78	0.06	47.8	33.5	18.8	1.03	117.7
Nanyishan	N105	2342.3-2346.6	N_2^1	0.75	1.05	19.4	34.6	46.0	0.96	124.2
	N14	4358-4457	E_3^2	1.32	1.74	13.3	49.4	37.4	0.84	—
	N2-5	—	E_3^2	1.67	0.31	19.5	50.4	30.1	1.06	128.0

$\Sigma DMCC_5 = \Sigma DMCC_5 \times 100/(n\text{-}C_7 + MCC_6 + \Sigma DMCC_5)$; $n\text{-}C_7 = n\text{-}C_7 \times 100/(n\text{-}C_7 + MCC_6 + \Sigma DMCC_5)$; $MCC_6 = MCC_6 \times 100/(n\text{-}C_7 + MCC_6 + \Sigma DMCC_5)$.
GTMRH: generation temperature of major reservoired hydrocarbons; $T = 140 + 15 \times \ln(2,4\text{-}DMC_5/2,3\text{-}DMC_5)$ (Mango [8]).

the ionization energy of the ion source was 86 eV. The experimental error determined by this analysis approach for gas yields was ±2% for each component.

Stable carbon isotope ratios were determined using a Finnigan MAT DELTAPlus mass spectrometer interfaced with a gas chromatograph. Gas components were separated on the gas chromatograph using helium as the carrier gas, converted into CO_2 in a combustion interface and then introduced into the mass spectrometer. Individual hydrocarbon gas components (C_1-C_5) and CO_2 were initially separated using a CP-CarboBOND column (50 m × 0.53 mm × 15 μm). The GC oven temperature was increased from 60°C to 200°C at 15°C/min (held 20 min). High purity methane (-28.5 ± 0.5‰, VPDB) is used as an internal standard for each sample test in order to test the stability and accuracy of the instrument. The precision was ±0.5‰. Stable carbon isotopic compositions were presented as $\delta^{13}C$ values relative to the VPDB scales.

3.3. Light Hydrocarbon Analysis. The light hydrocarbon composition was analyzed on an Agilent 6890N gas chromatograph (GC) with a 5973N mass spectrometer (MS). The GC was equipped with a split/splitless injector, a fused silica column (100 m × 0.25 mm i.d.× 0.5 μm film thickness), and a flame ionization detector (300°C). The injected sample volume was 1 ml (split ratio set to 20 : 1) and the injector temperature was 150°C. The oven temperature program was programed from 40°C (15 min) to 120°C (held 20 min) at 2°C/min and then to 290°C (held 20 min) at 12°C/min. Helium was used as the carrier gas (flow rate 1.0 ml/min). The MS conditions were electron ionization at 70 eV with an ion source temperature of 230°C. Compounds were

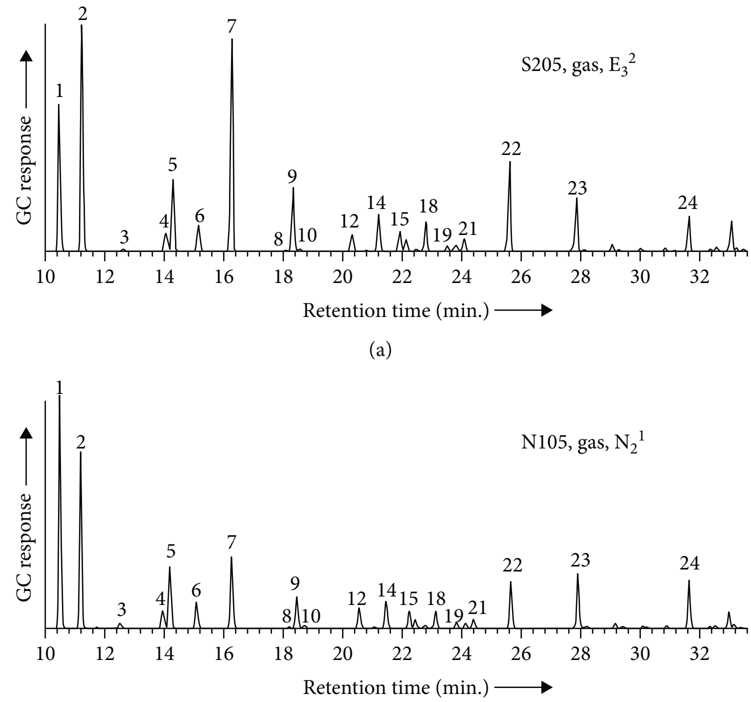

FIGURE 3: Representative gas chromatogram of light hydrocarbons (C_5-C_7) in natural gas from the western Qaidam Basin. The numbers and the corresponding light hydrocarbons are listed in Table 2.

identified by matching mass spectra with the NIST library of standard compounds (Figure 3 and Table 2). The used light hydrocarbons were obtained from integrated peak areas on the gas chromatograms.

4. Results

4.1. Natural Gas Components. The composition of natural gas in the western Qaidam Basin is mainly composed of hydrocarbons (average 91.6%). Nonhydrocarbon gases include nitrogen (average 4.2%), carbon dioxide (average 1.2%), and hydrogen sulfide (gas content in well S58 is 1.74%). Among the hydrocarbons, methane content is 58.7-95.6%, with an average of 81.0%; ethane content is 0.2-17.4%, with an average of 6.9%; and propane content is 0.1-9.4%, with an average of 1.9%. The gas dryness coefficient ($C_1/C_{1\text{-}5}$) is in the range of 0.66-0.99, with an average of 0.88, indicating that gases are mainly wet gases (Table 3).

4.2. Carbon Isotopes of Natural Gas. In the western Qaidam Basin, $\delta^{13}C_{methane}$ values range from -54.6‰ to -28.6‰, with an average of -38.9‰; $\delta^{13}C_{ethane}$ values range from -35.9‰ to -20.5‰, with an average of -26.9‰; and $\delta^{13}C_{propane}$ values range from -28.1‰ to -20.7‰, with an average of -24.3‰ (Table 3). Almost all gases from the western Qaidam Basin have an expected carbon isotopic distribution pattern among the C_1-C_3 alkanes, i.e., $\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3$, suggesting that most natural gases are primary gases. Only the N14 gas sample from the Nanyishan field has partial carbon isotopic reversal between ethane and propane ($\delta^{13}C_2 > \delta^{13}C_3$), probably implying admixture of gases from various origins.

TABLE 2: Number and the corresponding light hydrocarbon.

Number	Chemical name	Abbreviation
1	*i*-C_5	*i*-C_5
2	*n*-C_5	*n*-C_5
3	2,2-Dimethylbutane	2,2-DMC_4
4	Cyclopentane	CC_5
5	2-Methylpentane	2-MC_5
6	3-Methylpentane	3-MC_5
7	*n*-C_6	*n*-C_6
8	2,2-Dimethylpentane	2,2-DMC_5
9	Methylcyclopentane	MCC_5
10	2,4-Dimethylpentane	2,4-DMC_5
11	2,2,3-Trimethylbutane	2,2,3-TMC_4
12	Benzene	Benz.
13	3,3-Dimethylpentane	3,3-DMC_5
14	Cyclohexane	CC_6
15	2-Methylhexane	2-MC_6
16	2,3-Dimethylpentane	2,3-DMC_5
17	1,1-Dimethylcyclopentane	1,1-$DMCC_5$
18	3-Methylhexane	3-MC_6
19	*cis*-1,3-Dimethylcyclopentane	1,c3-$DMCC_5$
20	*trans*-1,3-Dimethylcyclopentane	1,t3-$DMCC_5$
21	*trans*-1,2-Dimethylcyclopentane	1,t2-$DMCC_5$
22	*n*-C_7	*n*-C_7
23	Methylcyclohexane	MCC_6
24	Toluene	Tol.

Table 3: Component and isotope properties of natural gas in the western Qaidam Basin.

Well	Main components (%)										Dryness	$C_1/(C_2+C_3)$	$\delta^{13}C$ (‰, VPDB)			R_o (%)*	R_o (%)**
	C_1	C_2	C_3	i-C_4	n-C_4	i-C_5	n-C_5	CO_2	N_2	H_2S			C_1	C_2	C_3		
S205	79.49	6.41	4.37	0.94	2.08	0.86	0.93	0.60	3.02	0.00	0.84	7.4	-40.6	-29.8	-26.4		0.8
S58	78.77	5.98	3.54	0.79	1.92	0.76	0.99	1.44	2.74	1.74	0.85	8.3	-40.0	-27.7			0.9
HN5-43-3	67.54	5.97	2.42	0.40	0.74	0.24	0.26	1.61	16.87	0.00	0.87	8.1	-54.6	-35.9	-28.1		0.6
YP3	72.94	10.97	0.89	1.09	0.02	0.33	0.31	1.24	5.35	0.00	0.84	6.2	-40.6	-27.9	-23.1		0.8
Y8-371	80.09	5.77	0.47	0.69	0.00	0.28	0.32	2.18	6.59	0.00	0.91	12.8	-42.1	-28.4	-26.8		0.8
YII5-13	91.01	1.75	0.50	0.38	0.01	0.17	0.24	1.82	1.71	0.00	0.97	40.5	-47.7	-28.2	-21.0		0.7
Y1-2-B1	73.34	9.91	1.00	1.24	0.01	0.31	0.25	1.70	4.33	0.00	0.85	6.7	-37.9	-27.2	-24.7		0.9
Y1-2-B4	87.93	4.27	0.38	0.48	0.00	0.17	0.16	0.68	2.85	0.00	0.94	18.9	-36.6	-26.8	-23.2		0.9
Y27-1-A6	86.30	4.95	0.43	0.58	0.00	0.20	0.20	0.75	3.20	0.00	0.93	16.0	-37.7	-26.7	-24.3		0.9
Y38-3-C	88.03	4.95	0.44	0.58	0.00	0.20	0.21	0.00	2.14	0.00	0.93	16.3	-39.5	-27.6	-25.0		0.9
Y29-1	81.66	6.29	0.60	0.74	0.01	0.19	0.19	1.38	4.10	0.00	0.91	11.8	-40.5	-30.9	-25.6		0.9
Y4-2	79.50	6.73	0.79	1.04	0.01	0.33	0.29	1.61	4.12	0.00	0.90	10.6	-39.6	-27.8	-24.4		0.9
Y9-4	81.61	3.59	0.60	0.79	0.01	0.28	0.26	0.70	7.22	0.00	0.94	19.5	-40.0	-28.7	-27.4		0.9
YD301	80.39	6.09	0.88	1.20	0.00	0.25	0.31	1.27	3.92	0.00	0.90	11.5	-39.5	-27.0	-23.9		0.9
YD3-3	79.96	6.63	0.68	1.01	0.01	0.41	0.40	1.78	4.37	0.00	0.90	10.9	-40.5	-26.8	-24.3		0.9
L4-5	80.76	7.14	0.64	0.89	0.01	0.31	0.42	1.15	3.64	0.00	0.90	10.4	-39.5	-27.0	-25.0		0.9
W8-11	87.32	6.20	2.51	0.41	0.60	0.17	0.17	0.09	2.17	0.00	0.90	10.0	-28.6	-24.3	-23.9	1.5	
WB6-19	86.33	6.28	0.62	0.72	0.01	0.21	0.28	0.00	1.42	0.00	0.91	12.5	-42.1	-26.1	-24.2		0.8
Z11-33-5	88.05	5.24	2.35	0.46	0.74	0.27	0.28	0.05	2.47	0.00	0.90	11.6	-34.8	-26.1	n.d.	1.0	
Z11-4-45	86.63	6.18	2.61	0.43	0.70	0.21	0.23	0.14	2.47	0.00	0.89	9.9	-33.5	-25.1	-23.4	1.1	
Z11-8-5	95.65	0.18	0.10	0.15	0.00	0.08	0.11	0.09	2.82	0.00	0.99	336.4	-39.1	-26.0	-25.2		0.9
Z218	67.97	14.23	7.12	1.20	2.06	0.60	0.58	2.15	3.58	0.00	0.73	3.2	-42.8	-29.4	-25.2		0.8
Z7-1-5	73.98	9.47	0.81	0.95	0.01	0.20	0.20	2.03	6.24	0.00	0.86	7.2	-42.0	-26.8	-23.6		0.8
Z7-2-3	80.87	9.62	3.33	0.48	0.80	0.21	0.23	0.71	3.47	0.00	0.85	6.2	-41.0	-27.2	-23.9		0.8
Z9-2-1	82.11	6.27	0.56	0.69	0.00	0.24	0.32	1.12	4.30	0.00	0.91	12.0	-39.2	-25.3	n.d.		0.9
ZP1	71.52	12.43	5.67	1.01	1.74	0.41	0.39	0.91	5.70	0.00	0.77	4.0	-39.4	-26.5	-23.5		0.9
ZT3	73.81	11.10	0.90	1.11	0.02	0.34	0.31	1.59	3.97	0.00	0.84	6.2	-36.8	-21.8	n.d.	0.8	
Q12-H5-11	58.66	17.43	9.41	1.23	2.08	0.40	0.35	0.66	8.70	0.00	0.66	2.2	-30.6	-25.9	-23.0	1.3	
N105	86.89	5.56	2.15	0.32	0.36	0.11	0.08	1.10	3.25	0.00	0.91	11.3	-33.0	-23.8	-21.8	1.1	
N14	90.31	2.12	0.16	0.00	0.01	0.00	0.01	6.08	1.22	0.00	0.98	39.6	-32.1	-20.5	-20.7	1.2	
N2-5	90.56	5.60	1.63	0.23	0.31	0.09	0.10	0.00	1.38	0.00	0.92	12.5	-34.3	-25.9	-23.8	1.0	

Note: dryness defined as $C_1/(C_1$-$C_5)$; n.d.: not determined. R_o (%)* values of humic type gases were calculated based on the $\delta^{13}C_1$-R_o relationship suggested by Dai and Qi [18], $\delta^{13}C_1 = 14.13 \times \log R_o - 34.39$; R_o (%)** values of sapropelic-type gases were calculated according to the $\delta^{13}C_1$-R_o relationship suggested by Shen et al. [19], $\delta^{13}C_1 = 40.49 \times \log R_o - 34.00$.

4.3. Light Hydrocarbon Composition. Light hydrocarbon compositions and relevant parameters are shown in Table 1. Representative chromatograms of light hydrocarbons (samples S205 and N105) are exhibited in Figure 3. Sample S205 has higher content of C_5-C_7 n-alkanes, and sample N105 has higher content of C_6-C_7 aromatics, implying different source rock kerogen types. Heptane and isoheptane values are commonly used to identify the maturity of gas and oil [3]. Due to the enhanced contents of alkanes in C_7 hydrocarbons from the saline lacustrine environment in the western Qaidam Basin [36], it is not suitable to study the maturity of gas by using heptane and isoheptane values.

A cross plot of n-C_7/MCC_6 and toluene/n-C_7 is often used to study the secondary alternation of petroleum in a reservoir [37]. Figure 4 shows that most gases in the western Qaidam Basin are in the area of the original, and some experienced secondary alternations. YII5-13, HN5-43-3, and Q12-H5-11 gas samples may have suffered biodegradation, and $\delta^{13}C_1$ values of YII5-13 and HN5-43-3 are -47.7‰ and -54.6‰, respectively, implying that these light hydrocarbons were from the bacterial decomposition of oil.

5. Discussion

5.1. Genetic Types of Natural Gases. A plot of $\delta^{13}C_1$ vs. $C_1/(C_2+C_3)$ is widely applied to identify kerogen types for gas generation [13, 38]. Figure 5 shows that most gases in the western Qaidam Basin are in the thermogenic area and a trace of them are close to the type-II kerogen area, some are distributed in or near the type-III kerogen area, and several fall

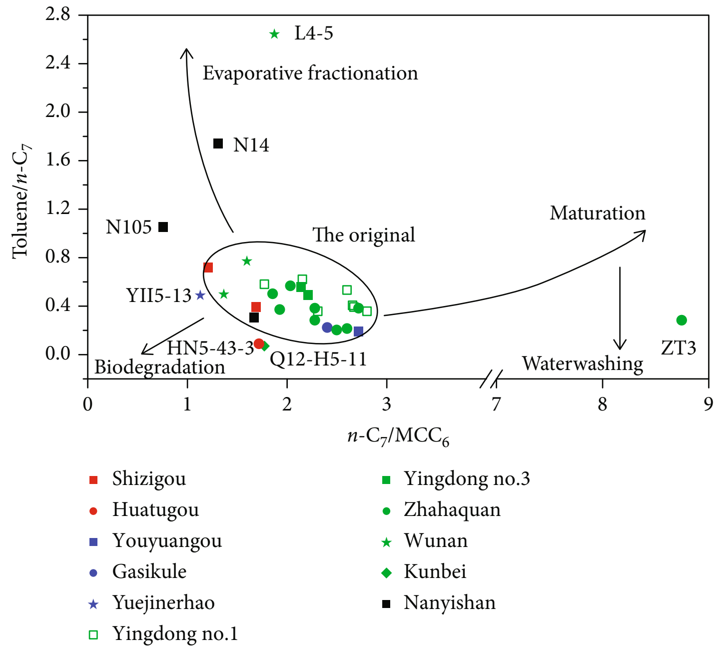

FIGURE 4: A cross plot of n-C_7/MCC_6 vs. toluene/n-C_7 of gases in the western Qaidam Basin (modified after [37]).

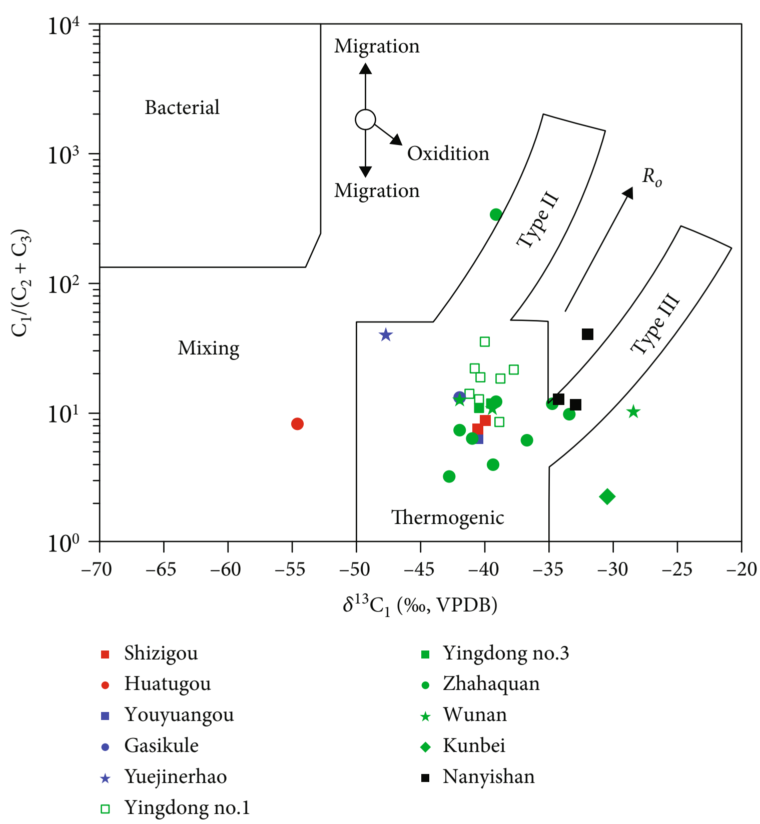

FIGURE 5: Diagram of $C_1/(C_2+C_3)$ vs. $\delta^{13}C_1$ of gases in the western Qaidam Basin (modified after [13, 38]).

in the area of mixing. The result indicates that most gases in the western Qaidam Basin were generated from sapropelic source rocks, while several gases in the Zhahaquan, Wunan, Kunbei, and Nanyishan fields were derived from humic source rocks, for example, W8-11, Z11-33-5, Z11-4-5, Q12-H5-11, N14, N105, and N2-5. Furthermore, $\delta^{13}C_2$ is often used as an identification of different types of gas. Zhang et al. [17] proposed a $\delta^{13}C_2 > -23.0‰$ cutoff for coal-type gas and a $\delta^{13}C_2 < -24.8‰$ cutoff for oil-type gas according to a research on the saline lacustrine natural gas in western Qaidam Basin. It is believed that most gases in the western Qaidam Basin are oil-type gases; W8-11, Z11-33-5, Z11-4-5, Q12-H5-11, ZT3, N14, and N105 are most likely the coal-type gases.

Relative abundance of *n*-heptane (n-C_7), methylcyclohexane (MCC_6), and dimethylcyclopentane ($DMCC_5$) may be used to identify the type of gases [6, 7]. Previous studies show that n-C_7 is predominantly derived from algae and bacteria; MCC_6 is primarily from lignin, cellulose, and components of higher plants; and $DMCC_5$ of various structures is mainly derived from the steroid and terpenoid compounds of aquatic organisms [2, 39]. Most gases in the western Qaidam Basin plot are in the sapropelic organic matter area, indicating that gases were mainly generated from sapropelic source rock (Figure 6). Only one gas sample, N105, falls in the humic organic matter area, implying that this gas was probably derived from humic source rock. Several gas samples have high contents of $\Sigma DMCC_5$, which attributes to strong depletion of n-C_7 from biodegradation.

Different genetic types obtained by C_1-C_3 fractions and C_7 fraction were observed in several gases in the Nanyishan and Zhahaquan fields of western Qaidam Basin. The $\delta^{13}C_1$ reveals that oil-type gas was generated at equivalent vitrinite reflectance (R_o) of 0.6-0.9% with an average of 0.8% and that coal-type gas was generated in the range of 0.8-1.5% with an average of 1.1%, which will be discussed below. Pyrolysis of source rocks shows that sapropelic kerogens are predominant in the Qaidam Basin, and also develops some humic kerogens [32, 40]. In contrast to the sapropelic source rock at the moderately mature stage, humic source rock at the late mature stage would generate more C_1-C_3 hydrocarbons and fewer C_7 hydrocarbons. Besides, a previous study proposed that coal-type gas in the Nanyishan field was derived from the coal measures of the Jurassic formation with a higher maturity [41], so there are probably contributions of coal-type gas from the Jurassic source rock in other fields in the western Qaidam Basin. Thus, it is inferred that mixed gas originated from the humic organic matter in deeply buried strata with an average R_o value of 1.1% and the sapropelic organic matter in the Tertiary deposition with an average R_o value of 0.8%.

Overall, based on the genetic type obtained from C_1-C_3 and C_7 hydrocarbons, four types of gases are identified: coal-type gas, featured by an enriched $\delta^{13}C_2$ value and high MCC_6 content; oil-type gas, characterized by a depleted $\delta^{13}C_2$ value and low MCC_6 content; biodegraded gas, featured by low n-C_7 content; and mixed gas, characterized by a relatively enriched $\delta^{13}C_2$ value and low MCC_6 content. Oil-type gas is the predominant-type gas in the western Qaidam Basin; coal-type gas is mainly distributed in the Zhahaquan and Nanyishan fields; mixed gas is mainly in the Zhahaquan, Wunan, and Nanyishan fields; and biodegraded gas is distributed in the Huatugou and Yuejinerhao fields.

5.2. Maturity of Natural Gas

5.2.1. Stable Carbon Isotope of Methane. Natural gas maturity is closely related to gas-source correlation, and maturity of source rock could be estimated with the empirical relationships between $\delta^{13}C_1$ and R_o [18, 19]. Because of different types of natural gases in the western Qaidam Basin, maturities of gases were calculated by the $\delta^{13}C_1$-R_o% relationships: Equation (1) for coal-type gas [18] and Equation (2) for oil-type gas [19].

$$\delta^{13}C_1 = 14.13 \log (R_o - 34.39), \tag{1}$$

$$\delta^{13}C_1 = 40.49 \log (R_o - 34.00). \tag{2}$$

Calculated thermal maturities of gases in the western Qaidam Basin are exhibited in Table 3. The R_o values for oil-type gases range from 0.6% to 0.9%, with an average of 0.8%, indicating that most gases are moderately mature, which matched well with the real thermal maturity of the Tertiary source rocks. The R_o values for coal-type gases are in the range of 0.8-1.5%, with an average of 1.1%, implying that coal-type gases reach mature and highly mature stages.

5.2.2. Chemical Composition of Heptane. The GTMRH obtained from light hydrocarbons was proposed by Mango [42] and modified by BeMent et al. [21] and Mango [8]. They suggested that the ratio of 2,4-dimethylpentane to 2,3-dimethylpentane (2,4-DMC_5/2,3-DMC_5) was a function of temperature, independent of time and kerogen types. The following empirically derived formula Equation (3) was published to calculate the GTMRH using C_7 light hydrocarbons [8].

$$T(^\circ C) = 140 + 15 \ln \left(\frac{2,4\text{-}DMC_5}{2,3\text{-}DMC_5} \right). \tag{3}$$

Based on the relationship, the GTMRH in the western Qaidam Basin are calculated and exhibited in Table 1. GTMRH positively correlated with the $\delta^{13}C_1$ and not significantly, probably because of the influence of the saline lacustrine sedimentary environment or the mixed sources from different organic matter. But gas samples in the Yingdong field show positive correlations between GTMRH and $\delta^{13}C_1$ which are most likely because they have same/similar sources (Figure 7(a)). Furthermore, GTMRH is positively correlated with ratio of 2-methylhexane to 3-methylhexane (2-MC_6/3-MC_6) which increases with increasing maturity (Figure 7(b)) [42].

GTMRH data reveal that the maturity of gases in the Yingdong, Zhahaquan, Wunan, and Nanyishan fields is higher than that in the Shizigou, Huatugou, Youyuangou, Gasikule, and Yuejinerhao fields (Figure 8). The $\delta^{13}C_1$ and gas dryness indicate higher maturity of gases in the former fields. Overall, maturity of gas increases from west to east in the southwestern Qaidam Basin. This is most likely because major source rocks near the Yingdong, Zhahaquan, and Nanyishan fields have higher maturity [43]. Moreover, most faults became increasingly active since the late Eocene, and these faults extend into the depression and form connections between the source rocks and the reservoir groups, which allow oil and gas with higher maturity to

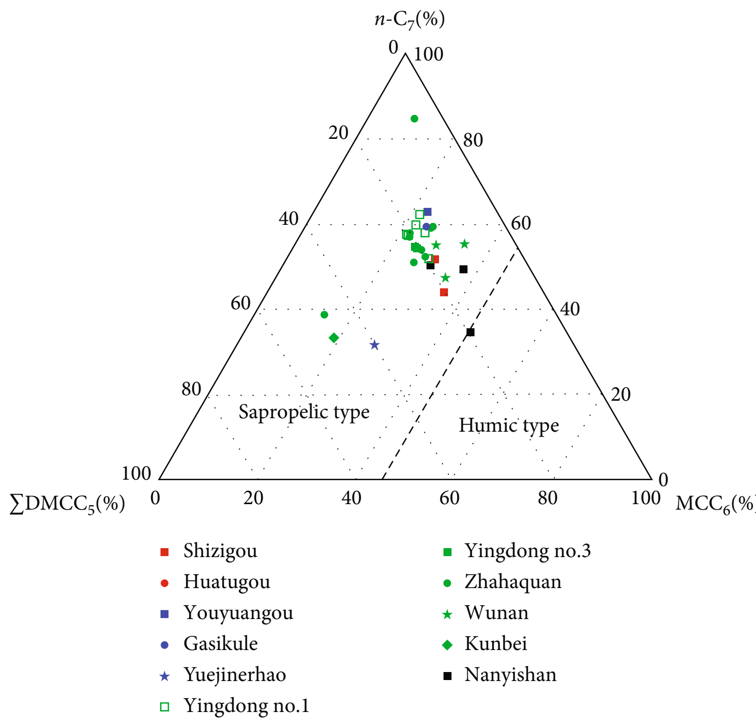

Figure 6: Ternary diagram of C_7 series in natural gases from the western Qaidam Basin.

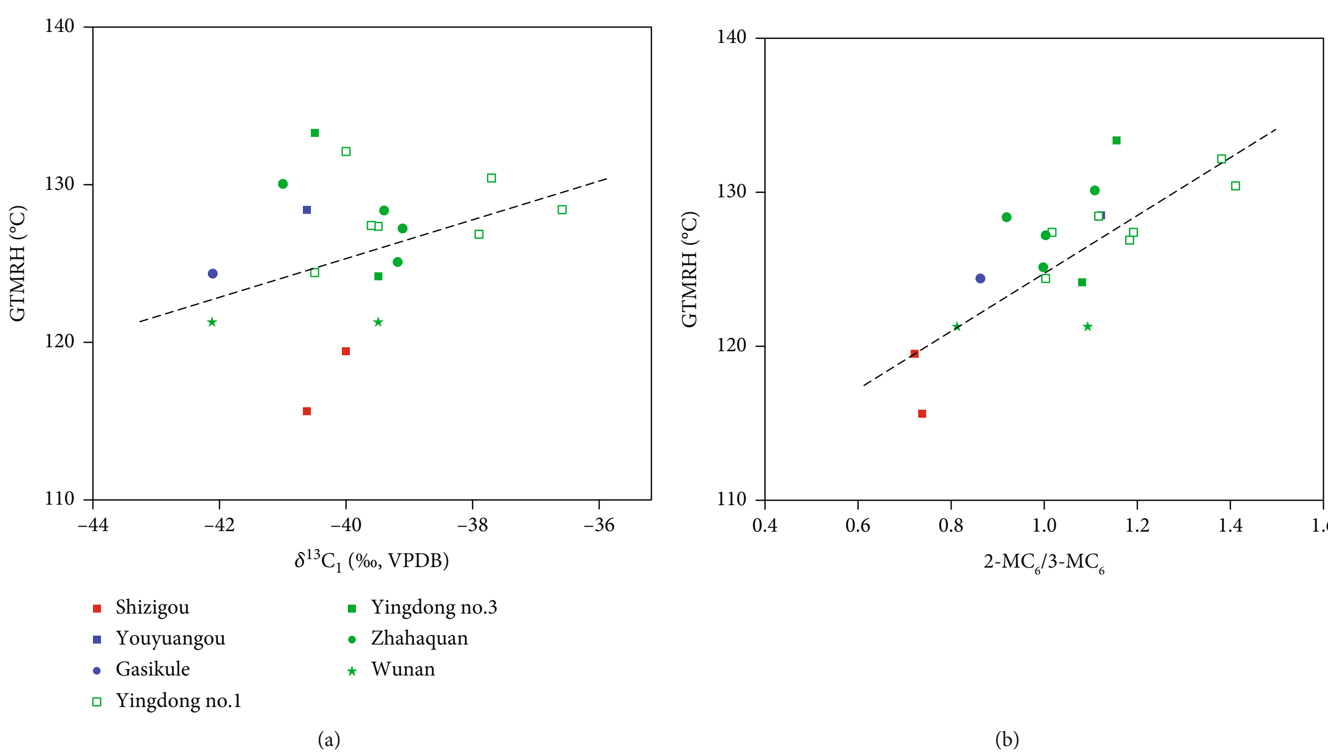

Figure 7: Cross plots of (a) GTMRH vs. $\delta^{13}C_1$ and (b) GTMRH vs. 2-MC_6/3-MC_6 for single sourced gases in the western Qaidam Basin.

migrate along the faults to reservoirs in midlevel and shallow formations [44]. Therefore, the maturity of gas in the Yingdong, Zhahaquan, Wunan, and Nanyishan fields is higher than that in others.

5.2.3. Application of Generation Temperature of Major Reservoired Hydrocarbons to Petroleum Charge History. Most studies suggest that there are primarily two petroleum charging episodes in the western Qaidam Basin [25–30]. The early

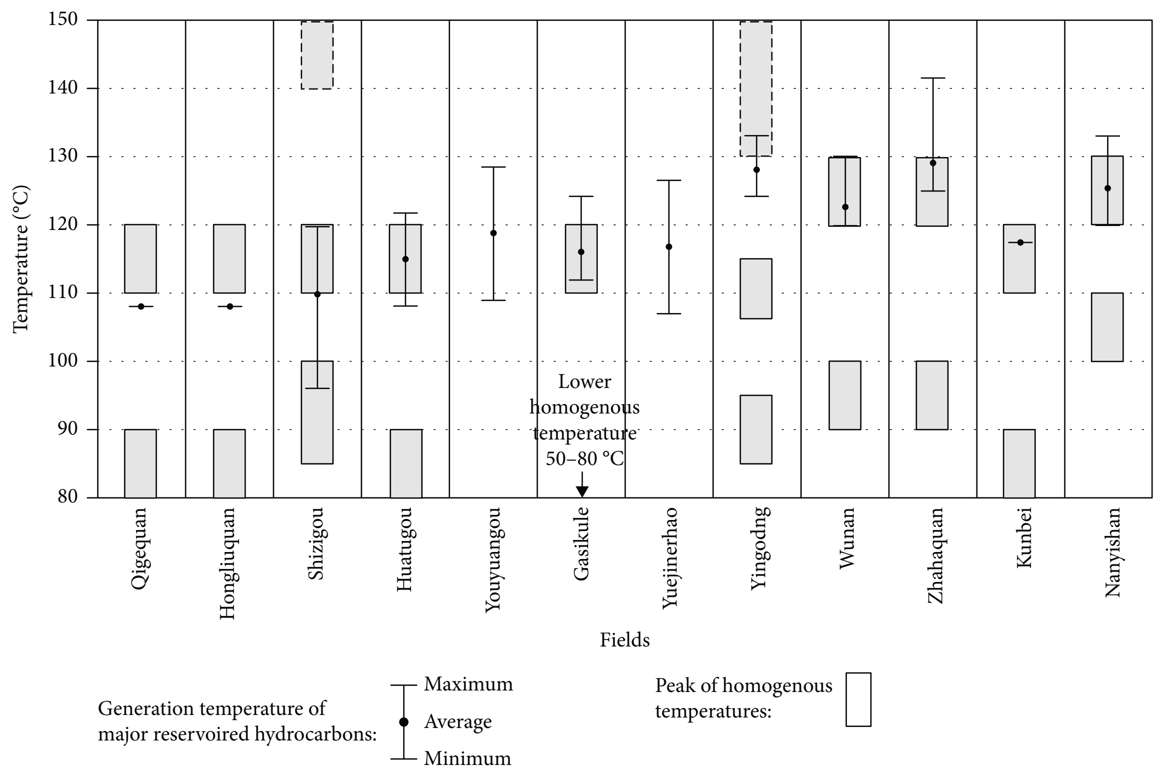

Figure 8: Relationship between generation temperature of major reservoired hydrocarbons (GTMRH) and peaks of homogenization temperature in different oil fields in the western Qaidam Basin. Supplementary GTMRH data for the Qigequan, Hongliuquan, Shizigou, Huatugou, Youyuangou, Gasikule, and Yuejinerhao oil fields are from Zhu et al. [31]. Peak homogenous temperature of fluid inclusion data for the Qigequan and Hongliuquan fields are from Fu et al. [1], Shizigou is from Li et al. [25], Huatugou is from Liu et al. [26], Gasikule and Nanyishan are from Li et al. [27], Yingdong is from Sui et al. [28], Zhahaquan is from Wang et al. [29], and Kunbei is from Chen et al. [30]. Solid border of the peak homogenous temperature is presented in published articles; the dotted border peak is based on latest studies.

charge occurred in the early Shangyoushashan period (about 10 Ma), and the later charge happened during the Shizigou period to the present (about 4-0 Ma), both of which matched well with the middle and late periods of the Himalayan movement [45]. In this study, all gas samples were collected from reservoired petroleum which was the result of effective hydrocarbon accumulation during the major charging episode. Hence, GTMRH obtained from the C_7 light hydrocarbon might be related to homogenous temperatures and could be used to study major petroleum charging episodes.

Combining with burial history (Figure 9), it shows that the early charge occurred in the early Shangyoushashan period with peak temperatures of 80-100°C, and the later charge happened during the Shizigou period to the present with peak temperatures of 110-130°C in the western Qaidam Basin (Figure 8). Both peak temperatures reveal that the maturity of petroleum in the Yingdong, Wunan, and Zhahaquan fields is higher than that in other fields in southwestern Qaidam Basin, which is in agreement with the study above. Figure 8 displays that GTMRH have a close relation with peak temperatures in the western Qaidam Basin. GTMRH in the Huatugou, Gasikule, Wunan, Zhahaquan, Kunbei, and Nanyishan fields overlap with higher peak temperatures, and their average GTMRH are in the range of higher ones, implying that the major petroleum charging episode is related to the later period. In the Qigequan and Hongliuquan fields, GTMRH are about 108°C, which are close to higher peak temperatures (110-120°C), indicating that the major petroleum charging episode is related to the later period, too. Though there is no homogenous temperature in the Youyuangou field, GTMRH indicates that petroleum was generated at a relatively higher mature stage. The GTMRH in the Shizigou field range from 96°C to 116°C, overlapping with lower and higher peak temperatures, and the average GTMRH value is more close to the higher one, suggesting that the major petroleum charging episode is related to the later period. Furthermore, the latest study has found higher homogeneous temperatures ranging from 140°C to 150°C in the Shizigou field. GTMRH in the Yingdong field are higher than both peak temperatures, and recent research suggested a petroleum charge event with homogeneous temperatures greater than 130°C. It may suggest an abundant amount of petroleum migration into shallow reservoirs due to strong tectonic activities during the late period of the Himalayan movement. On the whole, the GTMRH obtained from the C_7 light hydrocarbon could reflect a major petroleum charging episode, implying that a later charge of petroleum is predominant in the western Qaidam Basin.

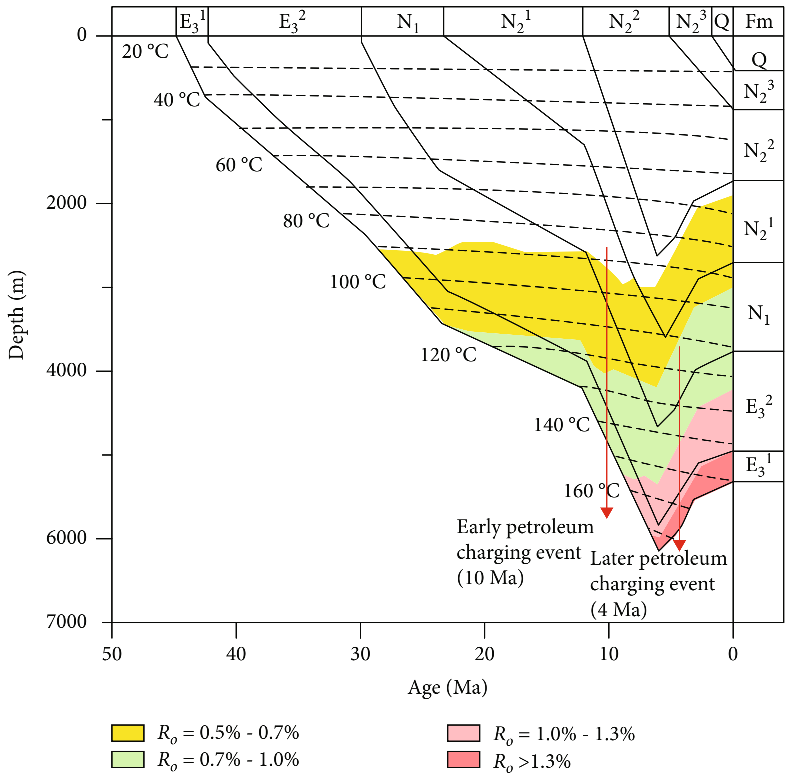

Figure 9: Sedimentary burial profile for the Yingdong field in the western Qaidam Basin (modified after [47]), and burial history is similar with most fields in the western Qaidam Basin.

The GTMRH calculated from the C_7 light hydrocarbon are mainly greater than 100°C, indicating that petroleum was mainly generated from the Tertiary source rocks at the middle stage of the oil window, consistent with the conclusion achieved from the $\delta^{13}C_1$ study. It is revealed from the light hydrocarbon study that if different molecular weight hydrocarbons in most reservoirs came from a similar origin, then petroleum in the western Qaidam Basin was primarily generated at the middle stage of the oil window and accumulated in the reservoirs during the late period of the Himalayan movement.

5.3. Implications for Hydrocarbon Exploration. The study above reveals that there are deep hydrocarbon fluid sources in the Shizigou, Yingdong, Zhahaquan, and Nanyishan fields. The GTMRH calculated from the C_7 light hydrocarbon indicates that petroleum accumulation mainly occurred during the late period of the Himalayan movement (about 4 Ma), simultaneous with the main oil-generation stage. Previous studies show that strong tectonic movement in the western Qaidam Basin leads to the formation of many deep faults and anticlinal traps [26, 44], which allows petroleum to migrate to traps along the faults. Thus, abundant petroleum accumulated in the midshallow layers and has been explored, and it is most likely to have a high exploration potential in deep formation. Besides, in the western Qaidam Basin, oil and gas were mainly derived from sapropelic source rocks of the Tertiary depositions and also some contributions of coal-type gas from the Tertiary and probably the Jurassic source rocks. In summary, deep reservoirs of paleouplifts adjacent to the hydrocarbon-generating depressions are estimated as a favorable area for further exploration in the western Qaidam Basin.

6. Conclusions

The gas composition of light hydrocarbons (C_5-C_7) and carbon isotopes was analyzed from 31 gases in the western Qaidam Basin. The genetic type and thermal maturity of natural gas were determined by C_1-C_3 and C_5-C_7 hydrocarbons. The generation temperature of major reservoired hydrocarbons was calculated from the C_7 light hydrocarbon and was applied to petroleum charge history. The following main conclusions have been reached:

(1) Four types of gases are identified: oil-type gas, coal-type gas, biodegraded gas, and mixed gas. The oil-type gas is the predominant-type gas in the western Qaidam Basin; coal-type gas is mainly distributed in the Zhahaquan and Nanyishan fields; mixed gas is mainly in the Zhahaquan, Wunan, and Nanyishan fields; and biodegraded gas is mainly distributed in the Huatugou and Yuejinerhao fields

(2) The R_o values of gas range from 0.6% to 1.5%, with an average value of 0.9%. The GTMRH calculated from the C_7 components range from 115.6°C to 141.7°C, with an average value of 126.5°C. Both maturity

indicators have relatively positive correlation and reveal that the maturity of gas increases from west to east in the southwestern Qaidam Basin

(3) The major petroleum charge episode in the western Qaidam Basin, which is during the late period of the Himalayan movement, is identified based on GTMRH combining with the homogenous temperature of petroleum inclusions

(4) Deep reservoirs of paleouplifts adjacent to the hydrocarbon-generating depressions are estimated as a favorable area for further exploration in the western Qaidam Basin

Data Availability

The experimental data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that there are no conflicts of interest.

Acknowledgments

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