Formation mechanism of condensates, waxy and heavy oils in the southern margin of Junggar Basin, NW China

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Abstract   It is a challenge to determine the source and genetic relationship of condensate, waxy and heavy oils in one given complicated petroliferous area, where developed multiple sets of source rocks with different maturity and various chemical features. The central part of southern margin of Junggar Basin, NW China is such an example where there are condensates, light oils, normal density oils, heavy crude oils and natural gases. The formation mechanism of condensates has been seriously debated for long time; however, no study has integrated it with genetic types of waxy and heavy oils. Taking the central part of southern margin of Junggar Basin as a case, this study employs geological and geochemical methods to determine the formation mechanism of condensates, waxy and heavy oils in a complicated petroliferous area, and reveals the causes and geochemical processes of the co-occurrence of different types of crude oils in this region. Based on detailed geochemical analyses of more than 40 normal crude oils, light oils, condensates and heavy oils, it is found that the condensates are dominated by low carbon number n-alkanes and enriched in light naphthenics and aromatic hydrocarbons. Heptane values of these condensates range from 19% to 21%, isoheptane values from 1.9 to 2.1, and toluene/n-heptane ratios from 1.5 to 2.0. The distribution of n-alkanes in the condensates presents a mirror image with high density waxy crude oils and heavy oils. Combined with the oil and gas-source correlations of the crude oils, condensates and natural gas, it is found that the condensates are product of evaporative fractionation and/or phase-controlled fractionation of reservoir crude oils which were derived from mature Cretaceous lacustrine source rocks in the relatively early stage. The waxy oils are the intermediate products of evaporative fractionation and/or phase-controlled fractionation of reservoir crude oils, while the heavy oils are *in-situ* residuals. Therefore, evaporative fractionation and/or phase-controlled fractionation would account for the formation of the condensate, light oil, waxy oil and heavy oil in the central part of southern margin of Junggar Basin, resulting in a great change of the content in terms of light alkanes, naphthenics and aromatics in condensates, followed by great uncertainties of toluene/n-heptane ratios due to migration and re-accumulation. The results suggest that the origin of the condensate cannot be simply concluded by its ratios of toluene/n-heptane and n-heptane/methylcyclohexane on the Thompson's cross-plot, it should be comprehensively determined by the aspects of geological background, thermal history of source rocks and petroleum generation, physical and chemical features of various crude oils and natural gas, vertical and lateral distribution of various crude oils in the study area.

Keywords   Condensate, Waxy oil, Heavy oil, Light hydrocarbons, Evaporative fractionation, Phase-controlled fractionation, Southern margin of Junggar Basin

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1. Introduction

Condensate is a kind of special crude oil that is usually composed of low carbon number hydrocarbons. The highly mature source rock (R_o 1.3–1.6%) can generate certain amount of highly mature condensate (Tissot et al., 1984), which has been proven by petroleum exploration in many petroliferous basins around the world. However, condensate is not necessarily the product of highly mature source rock. For example, type III organic matter and resin in coal could also generate condensate oil at low-mature stage (Snowdon, 1982, 1991; Boreham et al., 1993; Lu, 1994; Chen, 1995). The sedimentary organic matter which is originated from bacteria or reformed by bacteria is also favorable to form condensate oil at low mature stage (Wang et al., 1995). Alternatively, evaporrative fractionation (Thompson, 1987, 1988, 2010, 2016; Zhang et al., 2000; Su et al., 2004) and phase-controlled fractionation (Larter et al., 1991; Curiale et al., 1996a, 1996b; Su et al., 2000; Huang et al., 2001) could also form condensate oil, waxy oil and heavy oil. In addition, oil cracking (Horsfield et al., 1992; Peters et al., 2005) or "steady-state catalytic reaction" (Mango, 1990, 1994, 1997) could form light hydrocarbons or condensate oil. Therefore, it is still a big challenge to determine the genetic type of condensates in one given petroliferous area where developed multiple sets of source rocks with different maturities and various chemical features.

A large body of publications showed successful applications of cross-plot of toluene/*n*-heptane versus *n*-heptane/methylcyclohexane proposed by Thompson (1987, 1988) to determine the origin of condensates (Walters, 1990; Dzou et al., 1993; Ma, 1995; Ma et al., 1995; Masterson et al., 2001; Fu et al., 2003; Liang et al., 2003; Chen et al., 2003; He et al., 2004; Zhang et al., 2010; Gong et al., 2014; Chen et al., 2015; Su et al., 2015), and it is evaporative fractionation that accounted for the high toluene/*n*-heptane ratio. However, field observations also showed that evaporrative fractionation can result in a low toluene/*n*-heptane ratio (Su et al., 2003; Zhu et al., 2003; Su et al., 2013). This is inconsistent with traditional view in terms of the light hydrocarbon compounds as main indices to determine the cause of condensate oil. This questioned whether the light hydrocarbons can still be effective to determine the genetic types of condensate oil, and how to correctly determine it?

There are five sets of potential source rocks with different depositional environments and maturity in the southern margin of Junggar Basin. Previous studies showed that the types of oil and gas found in Horgos, Manas, Hutubi and other anticlines in the central part (Figure 1) are extremely complicated with normal black crude oil, brown light oil, yellow to colorless condensates, as well as black viscous heavy oil and a large number of natural gases, belonging to one typical complex petroliferous area (Wang et al., 2013; Chen et al., 2015a, 2015b). The natural gas in the anticlines of the southern margin of Junggar Basin was considered to be mainly coal-type wet gas or dry gas derived from the highly mature Jurassic coal-bearing source rocks (Wang et al., 2004[1], 2013; Chen et al., 2004; Liao et al., 2006, 2011; Liu et al., 2016). However, for the source of condensate oil, serious debates were received from the Chinese petroleum geochemists, regardless the causes of waxy crude oil and heavy oil. Using the methylcyclohexane content in the light hydrocarbons of the condensates and the Mango's classification plot of light hydrocarbons, Chen et al. (2004) suggested that the condensate oil in the Hutubi anticline was humic in origin and derived from the highly mature Jurassic coal-bearing source rocks. Jiang et al. (2006)[2] proposed that the condensates in the Manas anticline were mainly derived from Jurassic source rocks, with some contributions from Cretaceous lacustrine source rocks. Liao et al. (2006, 2011), Chen et al. (2008[3], 2016a, 2016b), Kang et al. (2008) and Wang et al. (2013) suggested that the crude oils in the Homatu anticlinal zone of the southern margin of Junggar Basin were derived from the Cretaceous lacustrine source rocks.

Why are the types of crude oils in the central part of southern margin of Junggar Basin diversified in respect to their physical and chemical properties? Are these condensates highly mature? Are these condensates derived from thermal degradation of highly mature source rock? How are waxy crude oil and heavy oil formed, and what is the relationship with the condensate?

In this study, a series of molecular geochemical analyses have been conducted on more than 40 condensates, crude oils and heavy oils from the southern margin of Junggar Basin. Integrated with previous oil-source correlation results (Chen et al., 2015b, 2016a, 2016b, 2016c) and the distributions of light hydrocarbons, the formation mechanism of condensate oil, waxy oil and heavy oil was discussed within the frame of regional tectonic evolution history and hydrocarbon generation history. The results could have implications for prediction of the origin of condensate oil.

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2.   Geological settings

2.1   Basic tectonic and structural units

The southern margin of Junggar Basin is 500 km long from east to west, and 40–60 km wide from north to south, with an area of 21000 km² with the Fukang fault zone as its east boundary and the Sikeshu sag as its west boundary. It is adjacent to the Shawan sag, Monan uplift, and Fukang sag of the Central Depression in the north, and the North Tianshan Mountains in the south. The southern margin of Junggar Basin belongs to the part of North Tianshan piedmont thrust belt structurally (Figure 1), and experienced three tectonic movements such as the Late Hercynian, the Indosinian-Yanshanian and the Himalayan. It is a first-order structural unit, and also the youngest and most complex fold belt in the Junggar Basin, featuring east-west segmentation and north-south zoning (Yang et al., 2004; Kuang et al., 2005, 2006; Chen et al., 2007; Guo et al., 2011). Based on the formation mechanism and structure pattern, the southern margin of Junggar Basin can be further divided into 4 second-order structural units: the Sikeshu sag, the Homatu anticline zone, the Qigu step-fault zone and the Fukang step-fault zone (Figure 1). Here we divide the southern margin of Junggar Basin into three parts for geochemical discussion: the west part (west of Dushanzi anticline), the central part from Dushanzi to Urumqi, and the east part (east of Urumqi).

2.2   Stratigraphic characteristics

The southern margin of Junggar Basin has continuously subsided since the Permian, and developed six sets of sedimentary strata (the Permian, Triassic, Jurassic, Cretaceous, Paleogene and Neogene-Quaternary) with the maximum deposition thickness of 15 km (general thickness of more than 10 km; Editor Group for Xinjiang Petroleum Geology, 1991; Wang et al., 2013). Details of these sedimentary strata are as following. The Lower Permian-the lower part of Middle Permian is mainly composed of a set of coarse clastic sediments. The Middle Permian Lucaogou Formation (P_2l) is a set of semi-deep to deep lake facies sediments with a thickness of 600–1600 m, and is usually rich in oil shale (Graham et al., 1990; Carroll et al., 1992). The Upper Permian is mainly fluvial facies-shallow lake facies-semi deep lake facies sediments with a thickness of 0–730 m. The Lower and Middle Triassic are mainly fluvial and shallow lake facies coarse clastic sediments with a thickness of 300–1300 m. The Upper Triassic is a set of delta, shallow lake, and semi-deep lake facies sediments with a thickness of 300–800 m, interbedded with swamp facies carbonaceous mudstones and thin coal layers. The Lower and Middle Jurassic is a set of laminated coal-bearing swamp facies and piedmont-fluvial facies sediments with the maximum thickness of nearly 3000 m. The Upper Jurassic is a set of foothill-fluvial facies coarse clastic sediments with a thickness of 0–350 m. The Lower Cretace-
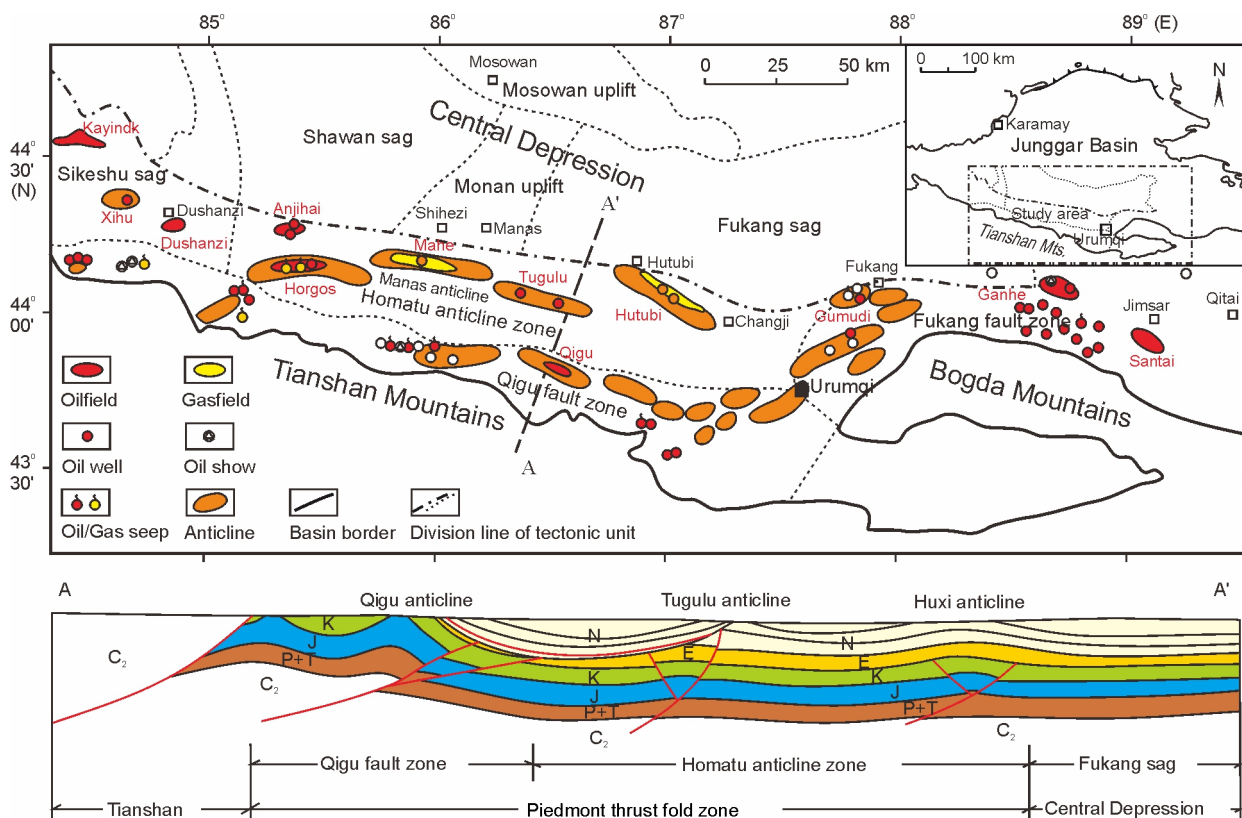

Figure 1   Structural traps and oil and gas field distribution in the southern margin of Junggar Basin.

ous is mainly a set of lacustrine facies mudstone sediments reaching 1600 m of thickness at maximum. The Upper Cretaceous is mainly piedmont fluvial facies of coarse clastic sediments with a thickness of 80–800 m. The Paleocene-Eocene is river-shallow lake facies sediments with a thickness of 15–855 m. The Eocene-Oligocene is relatively stable shallow lake-deep lake facies sediments with a thickness of 44–800 m, and the Oligocene-Pliocene is mainly shallow lake and fluvial facies sediments.

2.3 Potential source rocks

There are 5 sets of source rocks from the Permian, Triassic, Jurassic, Cretaceous and Paleogene in the southern margin of Junggar Basin. The central part of southern margin of Junggar Basin contains all 5 sets of source rocks, while the east and the west parts have 2 and 4 sets (Chen et al., 2008[3]), 2015a, 2016c; Wang et al., 2013). The Permian, Triassic, and Jurassic source rocks were buried up to 8–15 km in the depression, and have reached highly to over mature stages, indicating that they were potential candidates to generate highly mature condensates upon geologically burial history. The Cretaceous source rocks are mainly in the peak oil window, rather than at a late stage of generating highly mature condensates. The Paleogene source rock in the southern margin of Junggar Basin is immature to low mature in most areas, only in the Sikeshu sag and the central part of the southern margin of Junggar Basin, has it entered the early stage of oil window, but there is no possibility to generate highly mature condensates.

3. Samples and experiments

3.1 Samples

A large quantity of condensate reserves has been found in Hutubi, Manas and Horgos anticlines in the central part of southern margin of Junggar Basin. The Hutubi anticline is located in the third-row structural belt of the southern margin of Junggar Basin (Figure 1), and is mainly composed of Neogene, Paleogene, Cretaceous and Jurassic strata. The reservoirs are mainly composed of sandstone from the Paleogene Ziniquanzi Formation and the Cretaceous Donggou Formation (Figure 2a). The Hutubi anticline oil and gas field is dominated by natural gas with a small quantity of condensates. The Manas anticline is located in the middle of the second-row structural belt of the southern margin of Junggar Basin, whereas the Tugulu anticline on the east and the Horgos anticline on the west (Figure 1). In the Manas anticline, many layers of commercial natural gas and condensates have been found in the Paleogene Ziniquanzi Formation (Figure 2b). There is also light oil pool in the Cretaceous Donggou Formation underneath the condensate reservoir. The Horgos anticline was drilled from the Taxihe (N_1t) and Shawan (N_1s) formations of the Neogene, the Anjihaihe ($E_{2-3}a$), and Ziniquanzi ($E_{1-2}z$) formations of the Paleogene to the Cretaceous Donggou Formation (K_2d) and was found to contain many layers of oil and gas (Figure 2c, d), including the normal-density crude oil, low-density condensate and light oil, high-density heavy oil, as well as natural gas. The oil samples (Table
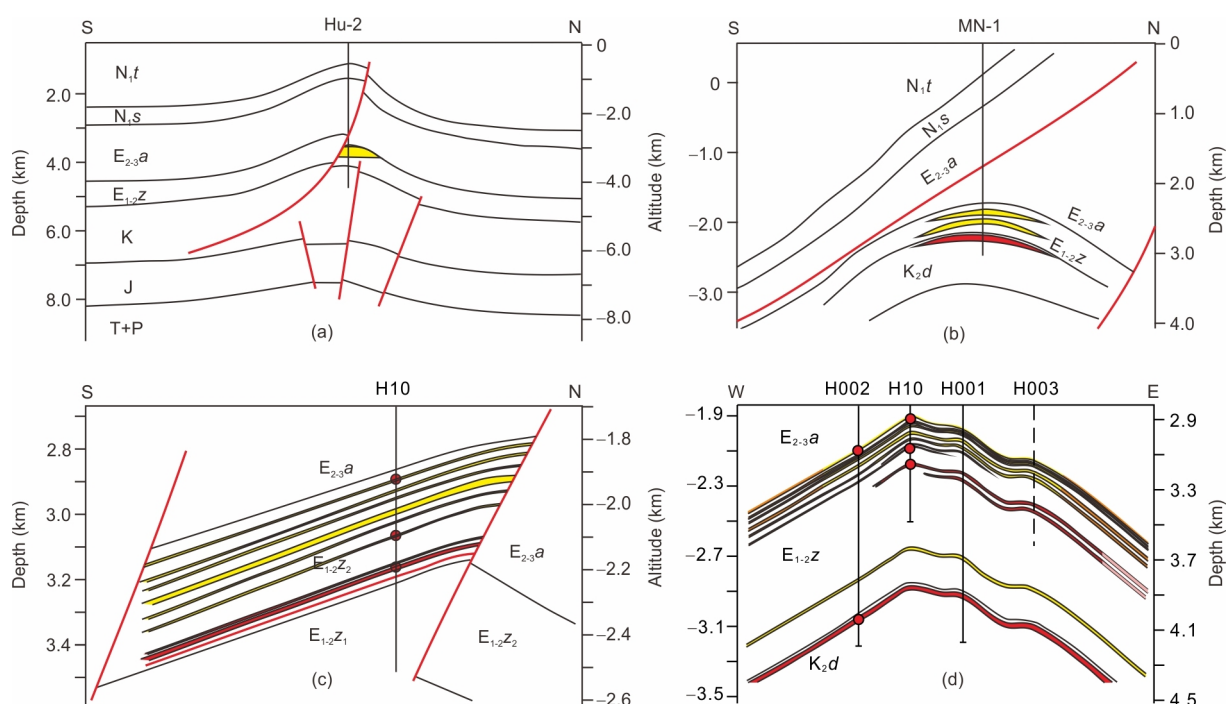

Figure 2 Sketch map showing oil and gas reservoir sections of anticline structure in the southern margin of Junggar Basin. (a) Hutubi anticline; (b) Manas anticline; (c) and (d) Horgos anticline (red spot for crude oil sampling). MN-1: Mana-1; H10: Huo-10; H001: Huo-001; H002: Huo-002; H003: Huo-003.

1) in this study were mainly collected from these anticlinal reservoirs in the southern margin of Junggar Basin.

3.2   Experiments

3.2.1   Fractional composition of condensate and crude oil

An IATROSCAN MK-6S Chromatography Analyzer was used, in accordance with the Standards of China Petroleum (SY/T 5119-2008), to quantify the compound-grouped fractions of the condensates and crude oils. Experiments were conducted at room temperature of 20–30°C and relative humidity of ≤ 65%. The reagents used were *n*-hexane, dichloroform, chloroform and isoamylalcohol. All reagents were purified before use except chromatography grade. The instrument was operated with air flux of 2000 mL/min, hydrogen flux of 160 mL/min and scanning speed 30 s/time.

3.2.2   Gas chromatography of light hydrocarbons and whole oil

The gas chromatography analyses of light hydrocarbons were performed on an Agilent 7890A with FID. A HP PONA capillary column (50 m × 0.20 mm i.d.; film thickness 0.5 μm) was used under a constant pressure mode, with N_2 as the carrier gas and a head pressure of 107 kPa, an average linear velocity of 14 cm/s, split ratio 50:1. The GC oven was initially set at 35°C, held for 10 min, subsequently programmed to 60°C at 0.5°C/min, and then from 60°C to 320°C at 6°C/min, and held at this temperature for 30 min. The equipment and conditions used for gas chromatography of whole oil were basically the same as that of light hydrocarbons except for programming process. The GC oven for whole oil was initially set at 35°C, held for 5 min, subsequently programmed to 320°C at 3.5°C/min, and held at this temperature for 30 min.

4.   Geochemical characteristics of condensate oil

4.1   Physical and chemical properties

The condensates from the Hutubi anticline are light yellow and colorless with the density of 0.7764–0.7832 g/cm³, viscosity around 0.8 mPa·s, freezing point from 20°C to 25°C, and the wax content of less than 2% (Table 1). Quantitative analyses of the compound-grouped fractions show that the saturated hydrocarbons range from 82% to 98%, the aromatics from 1% to 9%, while the resins and asphaltenes are particularly low. The condensates from the Manas anticline are yellow or light yellow with the density of 0.7612–0.7727 g/cm³, viscosity of 1.0–1.5 mPa·s, and freezing point below 20°C. The relative content of saturated hydrocarbons is between 91% and 96%, and the aromatic hydrocarbons from 3% to 8%, whereas the relative content of resins and asphaltenes

is less than 5%. The Horgos anticline has normal crude oil, light oil, condensate and heavy oil. The light oil has a density of 0.79–0.82 g/cm³, a viscosity of 1–5 mPa·s, a freezing point of 3–20°C, a wax content of 5–10%, a saturated hydrocarbon content of more than 80%, and aromatics content around 10%.

4.2   Gas chromatographic characteristics

The gas chromatograms of the whole condensates from the central part of the southern margin of Junggar Basin demonstrate that they are mainly composed of low molecular weight *n*-alkanes, the main peak carbon is nC_7 to nC_{10}, and the ratio of C_{21}^-/C_{22}^+ is basically >9.0 (Table 1, Figure 3). It should be noted that, due to the volatilization of low molecular weight hydrocarbons, the actual peak carbon of the condensates could be lower than what observed at nC_7. Although it is difficult to recover the volatized loss of low molecular weight hydrocarbons, these condensates are still rich in moderate molecular weight *n*-alkanes. For example, the condensate oil from the 3594–3597 m interval of Well Hu-2 in the Hutubi anticline has the highest number of *n*-alkanes up to nC_{20} or more despite the C_{21}^-/C_{22}^+ ratio is as high as 79.52. The condensate oil from the 3561–3575 m interval of Well Hu-2 has a carbon chain of *n*-alkanes up to more than nC_{25}. The condensates from the Manas anticline are very similar to that of Well Hu-2, that is, the relative content $>nC_{15}$ alkane accounts for a large proportion and the highest carbon number of *n*-alkane can be up to nC_{25} or more (Figure 3). In general, highly mature condensate is dominated by low molecular weight hydrocarbons, and usually with very low content of high molecular weight *n*-alkanes. The distribution of *n*-alkanes in the whole oil gas chromatogram indicates that the maturity of the condensate in the southern margin of Junggar Basin is not particularly high, and probably they should not be generated at the high maturity stage of the organic matter.

4.3   Composition of light hydrocarbons and maturity measurements

The distributions of light hydrocarbons in condensates from the Hutubi, Manas and other anticlines in the southern margin of Junggar Basin reveal that the light hydrocarbon composition before nC_8 is rich in paraffins, naphthenics and aromatics (Figure 4 and Table 2). All condensates have the highest content of methylcyclohexane (MCH), followed by nC_6 to nC_8 alkanes, cyclohexane, benzene and toluene. The content of other branched alkanes or naphthenics is relatively low. In the C_7 hydrocarbon compositions, the content of paraffins is 28–34% (15–17% of *n*-heptane, 13–17% of other branched alkanes), naphthenics is 39–46% (32–38% of methylcyclohexane, 6–7% of dimethylcyclopentane (DMCP), <2% of ethylcyclopentane), toluene is 22–31%.

Table 1 Basic physicochemical properties and geochemical parameters of some condensate and crude oils in the southern margin of Junggar Basin[a]

No.	Struc-ture	Well	Depth (m)	Horizon	Color/type	Den (g/cm³)	Vis (mPa·s)	Freez (°C)	Wax (%)	Sat (%)	Aro (%)	Res (%)	Asph (%)	CPI	MC	CC
6026	Hutubi	Hu-2	3561.5–3575.0	$E_{1-2}z$	Light yellow/condensate	0.7832	0.83	−26.2	3.51	82.25	9.06	4.41	4.28	1.15	C_{10}	9.28
5567	Hutubi	Hu-2	3594.0–3597	$E_{1-2}z$	Light yellow/condensate	0.7768	0.84	−20	1.36	97.43	1.28	0.64	0.21	1.33	C_8	79.52
0122	Hutubi	H001	3584–3590	$E_{1-2}z$	Colorless/con-densate	0.7764	0.82	−19	1.64	93.42	3.04	3.54	0.00	1.41	C_{10}	10.11
0123	Hutubi	H002	3536–3571.5	$E_{1-2}z$	Colorless/con-densate	–	–	–	–	92.48	3.35	4.17	0.00	1.29	C_9	10.27
0165	Manas	MN-1	2414–2446.5	$E_{1-2}z$	Light yellow/condensate	0.7612	1.02	−25.2	0.74	92.2	7.78	0.00	0.00	1.23	C_8	10.42
2884	Manas	MN-001	2365.51–2485	$E_{1-2}z$	Light yellow/condensate	0.7727	1.42	−26.1	0.50	91.06	4.53	3.15	1.26	1.21	C_{11}	9.88
7723	Manas	MN-001	2516–2522	$E_{1-2}z$	Light yellow/condensate	0.7652	1.38	−21.3	1.96	95.37	3.46	1.17	0.00	1.07	C_8	13.59
7724	Manas	MN-001	2787–2791	K_2d	Tawny/crude oil	0.7710	1.65	−15.4	2.25	88.74	8.89	2.15	0.22	1.10	C_{23}	1.53
5558	Horgos	H10	3064.0–3067.0	$E_{1-2}z$	Brownish lack/light oil	0.7967	1.10	10	6.62	87.38	8.54	2.73	1.33	1.14	C_8	7.10
5559	Horgos	H10	3159.0–3170.0	$E_{1-2}z$	Brownish black/crude oil	0.8303	7.36	35	12.57	91.95	4.54	2.89	0.61	1.13	C_8	1.55
5560	Horgos	H002	3097–3110	$E_{1-2}z$	Black/heavy oil	0.9409	48.00	37	11.57	40.02	33.49	12.47	14.01	1.08	C_{14}	1.71
5561	Horgos	H002	4050–4057	K_2d	Tawny/crude oil	0.8361	12.25	30	8.9	89.35	5.78	3.57	1.28	1.11	C_{14}	1.85
5562	Horgos	HQ1	644.0–645.0	N_1t	Brownish black/crude oil	0.8928	31.34	26	–	63.25	14.89	18.25	3.61	1.09	C_{27}	0.64
5563	Tugulu	TG-1	1840–1855	$E_{1-2}z$	Brown/crude oil	0.8619	11.06	33	–	93.66	3.61	2.11	0.61	1.11	C_{14}	1.85
5564	Tugulu	TG-2	1507.5–1570	$E_{1-2}z$	Brown/crude oil	0.8611	10.64	32	16.81	86.65	11.91	1.42	0.00	1.09	C_{23}	1.23
5565	Tugulu	TG-2	1786	$E_{1-2}z$	Brown/crude oil	0.8668	12.50	33	17.84	90.56	8.51	0.92	0.00	1.07	C_{23}	1.17
5566	Tugulu	Tu-002	1610–1625	$E_{1-2}z$	Brown/light oil	0.8345	3.44	20	6.55	91.21	7.04	1.59	0.16	1.16	C_{22}	1.26
6025	Qigu	Qi-8	2715–2737	T_1	Brownish black/crude oil	0.8806	37.78	10	9.02	62.61	8.03	15.97	13.39	1.20	C_{17}	2.14
6020	Gumudi	Mu-5	1300–1700	J_1s	Brown/crude oil	0.8456	5.82	11.5	13.93	82.36	8.18	6.82	2.64	1.20	C_{19}	2.57

a) Den: Density; Vis: Viscosity; Freez: Freezing point; Wax: Wax content; Sat: Saturated hydrocarbons; Aro: Aromatic hydrocarbons; Res: Resins; Asph: Asphaltenes; CPI: carbon preference index; MC: Main carbon; CC: C_{21}^-/C_{22}^+ ratio; H001: Huo-001; H002: Huo-002; MN-1: Mana-1; MN-001: Mana-001; H10: Huo-10; HQ1: Huoqian-1; TG-1: Tugu-1; TG-2: Tugu-2; –: not detected.
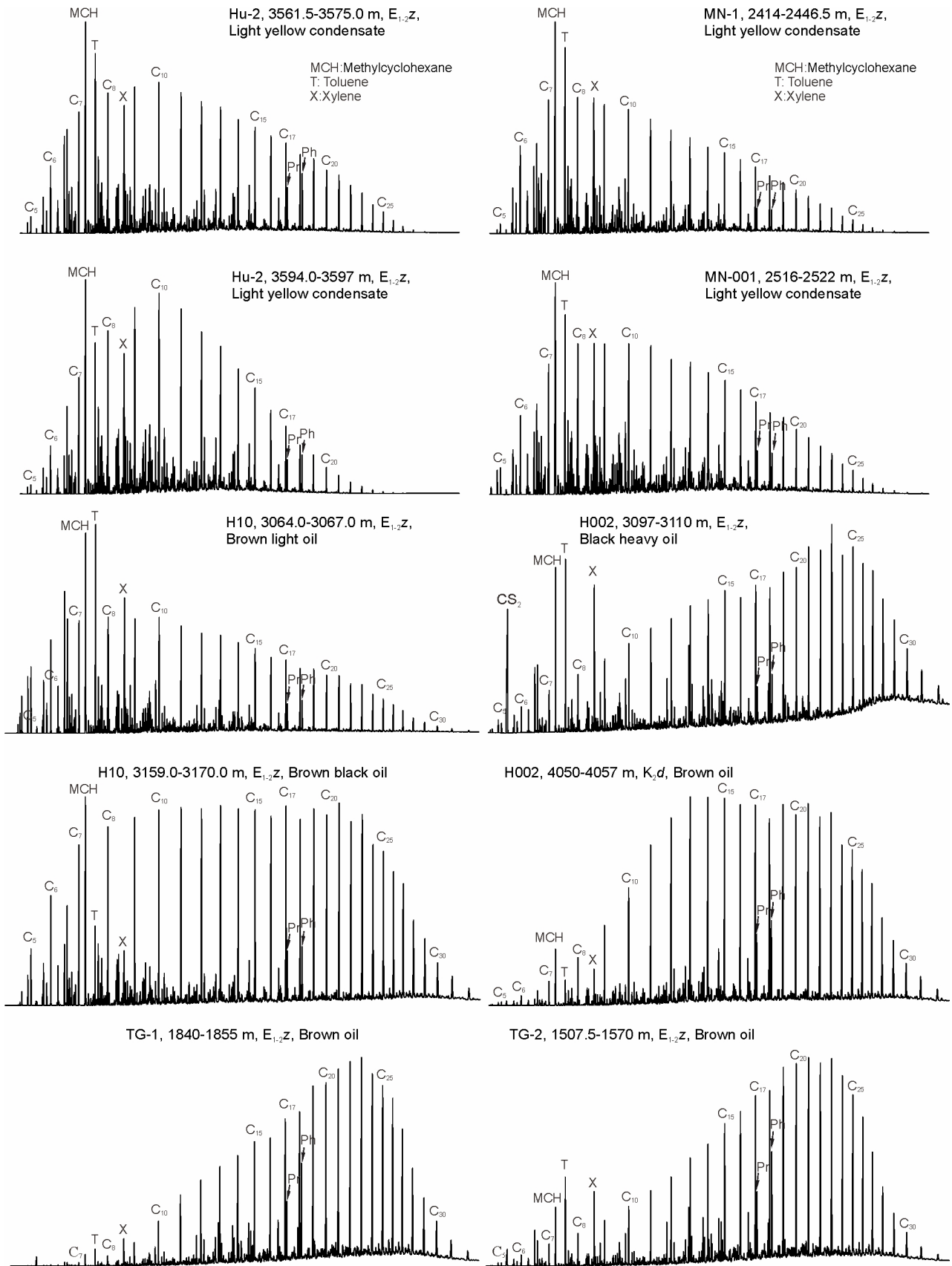
Figure 3   Gas chromatogram of whole oil of some condensate and crude oils in the central part of the southern margin of Junggar Basin.
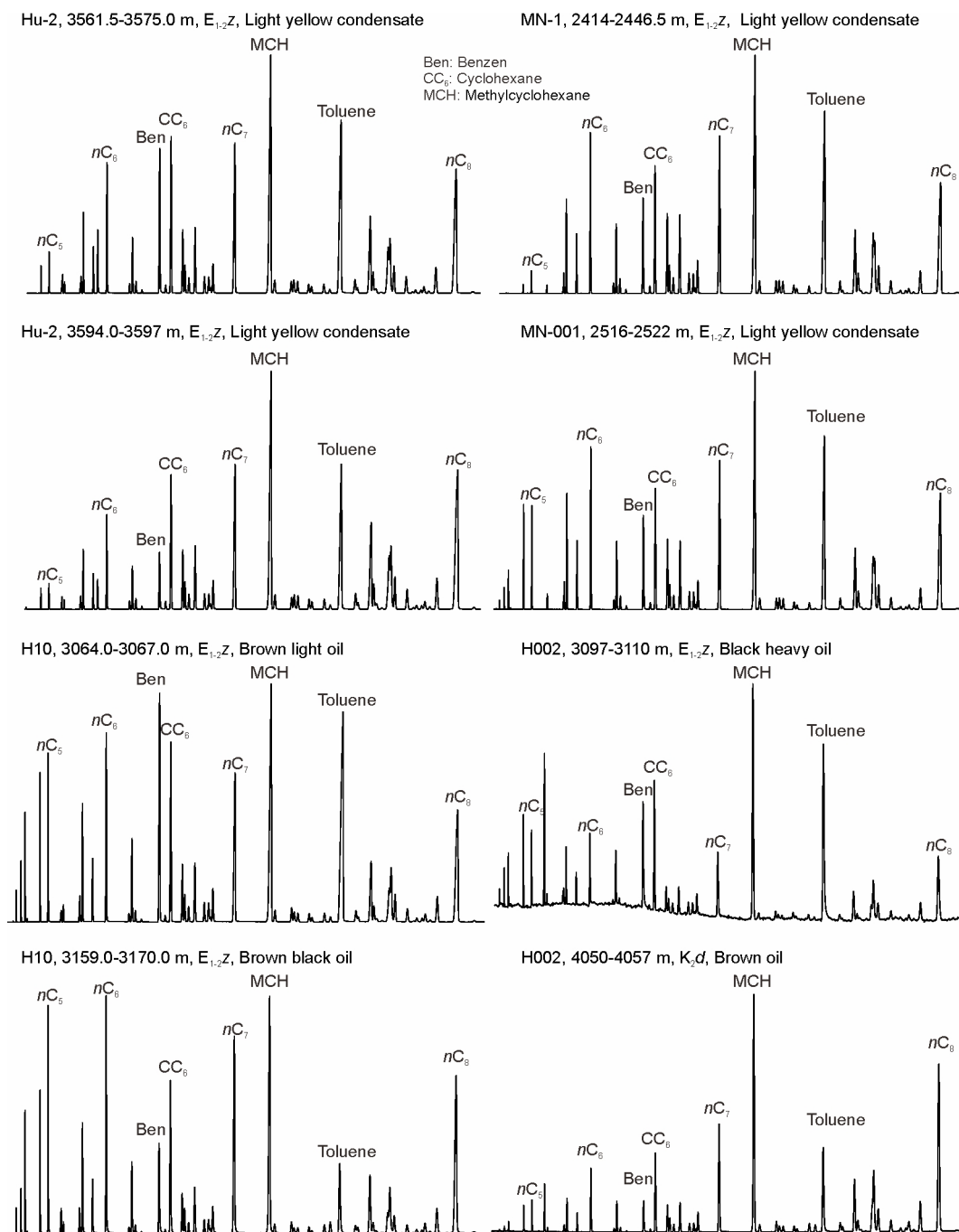
Figure 4 Gas chromatogram of light hydrocarbon of some condensate and crude oils in the central part of the southern margin of Junggar Basin.

Based on a large data set of light hydrocarbons from large numbers of crude oils and source rocks around the world, Thompson (1979, 1983) concluded that the relative content of C_7 alkanes with different structures can reflect the maturity of crude oil, and proposed two indices to determine the maturity, i.e., isoheptane and heptane values (Paraffin index 1 and Paraffin index 2). Crude oil with heptane value of 18–22% and the isoheptane value of 0.8–1.2 is paraffinic normal oils; oil with the heptane value of 22–30% and isoheptane value of 1.2–2.0 is mature oil; oil with heptane value of 30–60% and

isoheptane value of 2.0–4.0 is highly mature oil; and oil with heptane value <18% and isoheptane <0.8 is biodegraded oil. It is important to note that the prerequisite to use the isoheptane and heptane values to identify maturity of crude oil is that the crude oil has not undergone a secondary alteration. Otherwise, the isoheptane and heptane values will be affected, and the identification of oil maturity will also be biased.

With statistical studies on compositions of light hydrocarbons in large numbers of crude oils and source rocks from petroliferous basins in China, Wang (2011) concluded that

Table 2   The parameters of light hydrocarbon composition of some condensate and crude oils in the southern margin of Junggar Basin[a)]

No.	Well	Depth (m)	Horizon	Heptane value	Isohep-tane value	nC$_7$/MCH	Toluene/ n-Heptane	C$_7$Alk (%)	C$_7$Cyc (%)	C$_7$Aro (%)	iC$_7$ (%)	nC$_7$ (%)	MCH (%)	DMCP (%)	ECP (%)
6026	Hu-2	3561.5–3575.0	E$_{1-2}z$	19.97	1.98	0.48	1.59	31.69	42.18	26.13	15.20	16.48	34.50	6.47	1.21
5567	Hu-2	3594.0–3597	E$_{1-2}z$	19.89	1.97	0.46	1.29	32.76	45.32	21.93	15.74	17.02	37.29	6.72	1.31
0122	H-001	3584–3590	E$_{1-2}z$	19.33	1.97	0.46	1.70	30.69	42.31	27.01	14.76	15.93	34.72	6.42	1.16
0123	H002	3536–3571.5	E$_{1-2}z$	19.24	1.95	0.45	2.10	28.54	40.14	31.32	13.62	14.92	33.00	6.03	1.11
0165	MN-1	2414–2446.5	E$_{1-2}z$	20.63	2.09	0.52	1.61	33.78	39.82	26.40	17.37	16.40	31.39	7.17	1.27
2884	MN-001	2365.51–2485	E$_{1-2}z$	20.25	2.11	0.50	1.54	33.80	41.07	25.14	17.50	16.29	32.62	7.13	1.32
7723	MN-001	2516–2522	E$_{1-2}z$	20.41	2.07	0.49	1.59	32.59	41.42	25.99	16.22	16.37	33.24	6.93	1.25
7724	MN-001	2787–2791	K$_2d$	16.09	2.33	0.35	1.84	31.20	44.54	24.25	18.06	13.15	38.04	6.21	0.30
5558	H10	3064.0–3067.0	E$_{1-2}z$	18.96	1.58	0.45	2.45	26.03	39.03	34.94	11.79	14.24	31.63	6.21	1.19
5559	H10	3159.0–3170.0	E$_{1-2}z$	26.90	1.51	0.75	0.42	42.66	45.95	11.39	15.39	27.27	36.34	7.92	1.69
5560	H002	3097–3110	E$_{1-2}z$	15.07	1.34	0.31	2.33	23.13	47.85	29.02	10.66	12.47	40.01	6.32	1.52
5561	H002	4050–4057	K$_2d$	19.89	1.45	0.41	0.99	29.92	52.32	17.76	12.03	17.89	43.66	6.78	1.89
5562	HQ1	644.0–645.0	N$_1t$	17.62	2.23	0.40	1.64	31.24	44.59	24.17	16.49	14.75	36.90	6.18	1.52
5563	TG-1	1840–1855	E$_{1-2}z$	12.41	1.47	0.24	4.96	18.56	40.99	40.45	10.40	8.16	34.25	5.38	1.37
5564	TG-2	1507.5–1570	E$_{1-2}z$	16.23	1.61	0.36	4.16	20.85	35.58	43.58	10.37	10.48	29.49	5.07	1.02
5565	TG-2	1786	E$_{1-2}z$	15.55	1.53	0.29	3.79	18.57	41.57	39.86	8.07	10.50	35.61	4.86	1.10
5566	Tu-002	1610–1625	E$_{1-2}z$	17.13	1.52	0.39	2.75	24.87	40.09	35.03	12.12	12.75	32.93	6.13	1.02
6020	Mu-5	1300–1700	J$_1s$	25.53	2.29	0.62	0.47	42.98	46.14	10.89	19.60	23.38	37.42	7.09	1.62

a) nC$_7$/MCH: nC$_7$/Methylcyclohexane (n-Heptane/Methylcyclohexane); C$_7$Alk: C$_7$ Alkanes; C$_7$Cyc: C$_7$ Cycloalkanes (Naphthenics); C$_7$Aro: C$_7$ Aromatics; DMCP: Dimethylcyclopentane; ECP: Ethylcyclopentane.

isoheptane and heptane values were influenced by organic matter type, evaporative fractionation, biodegradation and other factors, and cannot accurately determinate the maturity of crude oil. Wang (2011) further proposed modified criteria after Thompson's (1983), and divided the crude oil into immature, low mature to mature and highly to over mature with the heptane value <18% and isoheptane value <1.0, the heptane value of 18–30% and isoheptane value of 1.0–2.0, and the heptane value>30% and isoheptane value >2.0, respectively.

In the Hutubi, Manas, Horgos and other anticlines in the central part of the southern margin of Junggar Basin, both the normal crude oil and condensate or heavy oil, the heptane and isoheptane values change within a relatively small range (Table 2), i.e., the heptane values range from 12.4% to 26.9%, and the isoheptane values are between 1.16 and 2.33. The heptane values of condensates range from 19% to 21%, and the isoheptane values are between 1.9 and 2.1. With the help of the criteria proposed by Thompson (1979, 1983) and Wang (2011), crude oil, condensate, or heavy oil in the central part of the southern margin of Junggar Basin, whether they have undergone a secondary alteration or not, are essentially mature oil rather than highly mature oil.

On the other hand, Mango (1987, 1990, 1994, 1997) and BeMent et al. (1995) suggested that the ratios of 2-methyl-cyclohexane/3-methylcyclohexane and 2,4-dimethylpen-tane/2,3-dimethylpentane (2,4-DMP/2,3-DMP) are function of temperature. These ratios correlate well with the temperature of hydrocarbon generation upon thermal maturation, whatever the age and type of organic matter, burial time and rate are. Mango (1997) proposed a formula to calculate the temperature: T=140+15ln(2,4-DMP/2,3-DMP). The ratios of 2,4-DMP/2,3-DMP in the condensates in the southern margin of Junggar Basin are basically between 0.33 and 0.50, and the maximum calculated temperatures range from 123°C to 130°C. However, most of the crude oil in this area has experienced severe secondary alterations, and the composition of the light hydrocarbons changed much compared to its originals (as discussed later). Therefore, the ratio of 2,4-DMP/2,3-DMP may not reflect its original value, and the calculated temperature cannot reveal the highest temperature reached by organic matter (Peters et al., 2005). However, the calculated temperature of crude oil from the 3157–3170 m interval in Well H10 (No. 5559 in Table 2, which is not subject to secondary alternation, discussed below) is only 120°C and is at the peak oil generation, suggesting that crude oil without secondary alteration in this region is products of organic matter at the peak oil generation.

In addition, our previous studies showed that the C_{29} sterane 20S/(20S+20R) values of crude oil, light oil and condensate in the Hutubi, Manas and Horgos anticlines are between 0.40 and 0.55, and the ββ/(αα+ββ) values range from 0.50 to 0.60 (Chen et al., 2015b, 2016c). For example, the 20S/(20S+20R)

and ββ/(αα+ββ) values for the condensate of Well Hu-2 are only 0.41 and 0.52 respectively. The maturity R_c calculated by the aromatics methylphenanthrene index (Radke, 1983, 1986, 1987) is only 0.42% (significantly low), and the maturity R_c calculated by the methylphenanthrene distribution fraction (Kvalheim et al., 1987) is only 0.87%.

Results from the light hydrocarbon composition and biomarker maturity parameters suggest that the condensates in the central part of the southern margin of Junggar Basin are mature condensates and are the products of the source rock at mature oil generation stage rather than highly mature stage.

5.   The formation mechanism of condensates, waxy and heavy oils

The physicochemical properties of the oil discovered in many anticlinal structures in the central part of the southern margin of Junggar Basin are extremely variable, including normal density crude oil, low density condensate and light oil, higher density waxy oil and high density heavy oil (Table 1). Furthermore, there is a significant amount of natural gas in these anticlines. The oil source correlation demonstrated that the carbon isotopic compositions of individual molecular hydrocarbons and biomarkers are very similar among normal crude oil, light oil, condensate oil and heavy oil, all of which were derived from the Cretaceous Qingshuihe Formation source rocks (Chen et al., 2016a, 2016b, 2016c). However, why did these crude oils exhibit large differences in basic physicochemical properties? In terms of the fact that large quantities of natural gases were found in these anticlines, the reformation of the reservoirs due to natural gas injection in the later stage may be the essential cause in respect to complicated physical and chemical properties of crude oil. This reformation is due to the "evaporative fractionation" proposed by Thompson (1987, 1988) and the "phase-controlled fractionation" proposed by Larter et al (1991).

5.1   Evaporative fractionation: The main formation mechanism

5.1.1   *Evidence from distribution of n-alkanes*

Thompson (1987) found significant fractionation of the crude oil by multiple injections of excess methane into a closed vessel containing crude oil. The low-carbon component is dissolved in the natural gas and migrated with the natural gas. With the increase of the fractionation degree, the carbon number of the fraction which is fractionated gradually increases, while the content of low-carbon hydrocarbons in the residual oil gradually decreases, and the relative content of the high-carbon number hydrocarbon gradually increases. With this, Thompson (1987, 1988) proposed the concept of "evaporative fractionation" to describe secondary alternation

of crude oil in reservoirs. Su et al. (2004) conducted similar simulations and obtained similar results.

As shown in Figure 3, the *n*-alkanes of condensates in the central part of the southern margin of Junggar Basin are basically the highest in the range of nC_7 to nC_{10}, followed by a decreasing trend with increasing carbon number. The low-carbon *n*-alkanes before C_{20} are usually the main constituents. The heavy oil from the 3097–3110 m interval of Well H002 is totally different from those of condensates, and it clearly shows that the relative content of high-carbon number *n*-alkanes is higher than that of the low-carbon number *n*-alkanes. The interesting thing is that the distribution of *n*-alkanes before C_{25} from this heavy oil can be a mirror image with condensate. Many crude oils from the southern margin of Junggar Basin share similar *n*-alkane distribution as that of the heavy oil from the Well H002, such as the oils in the Cretaceous Donggou Formation of 4050–4057 m interval of Well H002, and TG-1 and TG-2 wells of the Tugulu anticline (Figure 3). These crude oils also have relatively high densities (Table 1). In contrast to these condensates and heavy oils, the distributions of low-carbon number and high-carbon number alkanes in crude oil from 3159–3170 m interval of Well H10 are well balanced and belong to the distribution pattern of normal crude oil. If the crude oil of Well H10 is taken as a reference, the changes of *n*-alkanes in the condensate and heavy oil in the central part of the southern margin of Junggar Basin are totally in agreement with the "evaporative fractionation" suggested by Thompson (1987). The condensate oil belongs to the low-molecular-weight light fraction which is carried out by the natural gas, termed "evaporative fractionation", while the heavy oil belongs to the residual oil. Therefore, condensate oil and heavy oil are two end products of evaporative fractionation. Losh et al. (2002, 2010) also observed a similar phenomenon in many reservoirs along the offshore Louisiana of the Gulf of Mexico basin, which was believed to be due to gas washing (evaporative fractionation), and calculated the maximum loss of *n*-alkanes up to 91%.

5.1.2 Evidence from aromatic distribution

Thompson (1987) showed that, with the development of evaporative fractionation, the content of low-carbon aromatic hydrocarbons in the liquid fraction increased gradually compared to the same carbon number of *n*-alkanes, and suggested that the genesis of petroleum enriched in light aromatics (benzene, toluene, and xylene), cycloalkanes (methylcyclopentane, cyclohexane and methylcyclohexane) may be related to evaporative fractionation, and the degree of evaporative fractionation of crude oil can be characterized with the toluene/*n*-heptane ratio (Figure 5).

A very distinct feature of condensates and light oil in the Hutubi, Manas and Horgos anticlines in the central part of the southern margin of Junggar Basin is that these crude oils are rich in benzene, toluene and xylene (Figure 3, Figure 4).
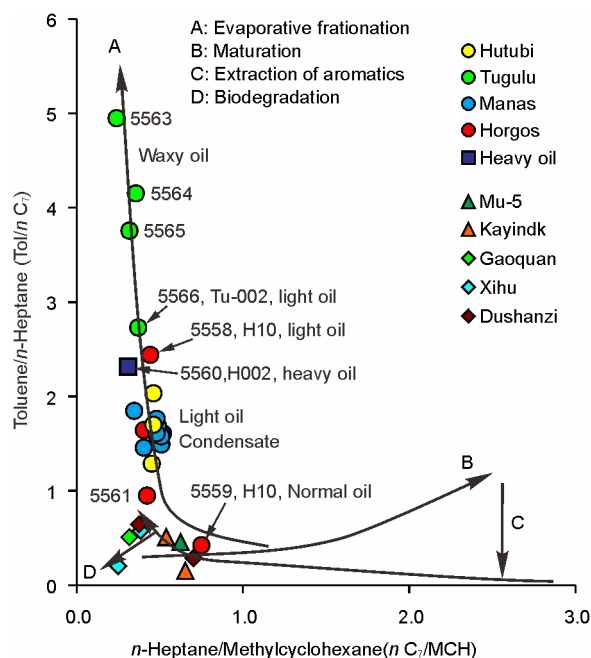

Figure 5   The cross-plot of paraffinicity versus aromaticity of the condensate, crude oils in the southern margin of Junggar Basin.

Toluene is the second peak compared to the highest peak of methylcyclohexane, usually higher than *n*-heptane, the toluene/*n*-heptane ratios are generally between 1.3 and 2.0 (Table 2, Figure 5), and the benzene/*n*-hexane ratios range from 0.7 to 1.6. However, the low-carbon number aromatics in the normal crude oil (No. 5559) from 3157–3170 m interval of Well H10 in the Horgos anticline is relatively low, and its toluene/*n*-heptane ratio is only 0.42 (Table 2, Figure 5). Clearly, the relative contents of low-carbon number aromatics in condensates and light oils from these anticlinal structures are higher than those of the normal oil from 3157–3170 m interval in Well H10. These condensates and light oils belong to the product of evaporative fractionation according to Thompson's (1987) cross-plot. On the other hand, some crude oils with high density or heavy oils in the central part of the southern margin of Junggar Basin also contain high contents of benzene and toluene, and the toluene/*n*-heptane ratio is even higher than those condensates. For example, the heavy oil from Well H002 has a toluene/*n*-heptane ratio of 2.33 (No. 5560 in Table 2, Figure 5); the waxy crude oils from TG-1 and TG-2, which contain wax content up to 16% (Table 1) and mainly is composed of saturated hydrocarbons and aromatics with resins plus asphaltenes <1.5%, have the toluene/*n*-heptane ratios as high as 4.16–4.96 (No. 5563, 5564, 5565 in Table 2, Figure 5). These heavy oils or waxy oils are usually lack of low carbon number *n*-alkanes and enriched in high carbon number *n*-alkanes (Figure 3), indicating residuals from the evaporative fractionation process or the product of the late stages of evaporative fractionation. Hence, the toluene/*n*-heptane ratio cannot properly determine the genetic types of condensates,

and light oil or waxy oil can be distilled off the product or residual product.

5.1.3 Discussion on evaporative fractionation

Using PVT simulation experiments and analyses of the light hydrocarbon compositions in condensates, Thompson (1987, 1988) found that excessive gas injection into the reservoir can lead to fractionation of the crude oil in the reservoir. The results showed that low-carbon number light hydrocarbons migrate out of the system with increasing natural gas injection and form condensed oil as the temperature/pressure decreases, while high-carbon number heavy hydrocarbons remain in the reservoir to form residual oil. The final result of the fractionation is the increased concentration of light aromatic and naphthenic hydrocarbons in the residual oil. Based on this finding, Thompson (1987, 1988) proposed a ratio of toluene/*n*-heptane as a measure of the degree of fractionation, and used the cross-plot of the toluene/*n*-heptane ratio versus the *n*-heptane/methylcyclohexane ratio to determine the genetic type of condensates. However, from the toluene/*n*-heptane ratios of all types of crude oils in the anticlines of the southern margin of Junggar Basin (Figure 5), the toluene/*n*-heptane ratio does not indicate the degree of fractionation of the crude oil, nor does it distinguish whether the condensate, light oil or waxy crude oil is a fractionated product or fractionated residue.

The evaporative fractionation by gas invasion is an extremely complicated physicochemical process. The composition of crude oil strongly depends on the processes involved, including the intrusion of gaseous hydrocarbons, the intermixing of gaseous hydrocarbons with liquid hydrocarbons, volume expansion, deasphalting, migration of gaseous or liquid hydrocarbons, desorption of gaseous hydrocarbons and precipitation of liquid hydrocarbons after temperature and pressure reduction, etc. The processes involved are still unclear to date. In the gas-liquid two-phase

system, the solubility of gaseous hydrocarbons in liquid hydrocarbons or the solubility of liquid hydrocarbons in gaseous hydrocarbons, the desorption of gaseous hydrocarbons, and the condensation of liquid hydrocarbons are functions of many factors as temperature, pressure, hydrocarbon polarity, fugacity of different hydrocarbon components (gas-liquid equilibrium constant), saturated vapor pressure etc. For example, the saturated vapor pressures of *n*-heptane, methylcyclohexane, and toluene at 22°C are 5.24, 5.33 and 3.33 kPa, respectively, and xylene is 0.74–0.98 kPa at 22°C. Based on the saturated vapor pressure of these compounds, methylcyclohexane and *n*-heptane could be more easily to become gas-phase fractions than toluene and xylene, then to be fractionated into natural gas under the same temperature and pressure condition.

From the viewpoint of mass balance and component-dependent fractionation principle, residual crude oil from evaporative fractionation should be relatively enriched in high carbon number *n*-alkanes and low molecular weight aromatics, with relatively less of low carbon number *n*-alkanes. On the contrary, the liquid components that migrate out of the system accompanying natural gas are enriched in low-carbon number *n*-alkanes and less high carbon number *n*-alkanes and low molecular weight aromatics. Theoretically, it is not a case that the aromatics in the condensate increased with enhancing fractionation, suggesting that evaporative fractionation cannot make the fractionated products and the residues enrich in light aromatics and naphthenics at the same time.

Thompson (1987) observed that low-carbon aromatics in the fractionated liquid condensate increased gradually with increasing fractionation in his experiment. In fact, this is only a reflection of the relative content of low-carbon number aromatics and low-carbon number *n*-alkanes in the products at each stage of the fractionation process rather than the accumulative products of the whole fractionation process. In other words, the products of the latter stage do not contain the prod-

Table 3 Analytical parameters, experimental oils and condensates[a]

Stage	Sample	B	X	F	H
0	Original oil	1.24	0.81	0.67	21.3
1	Condensate 1	0.85	0.80	0.77	22.2
2	Condensate 2	1.14	0.89	0.69	20.8
3	Condensate 3	1.33	1.05	0.62	20.3
4	Condensate 4	1.77	1.21	0.46	17.6
5	Condensate 5	2.24	1.40	0.45	17.9
6	Condensate 6	3.18	1.79	0.35	15.1
7	Condensate 7	4.50	2.25	0.29	13.2
8	Condensate 8	6.13	2.70	0.25	11.9
9	Condensate 9	8.00	3.72	0.20	10.7
10	Condensate 10	8.50	4.63	0.21	10.3
11	Condensate 11	7.00	10.14	0.20	10.0

a) Modified from Thompson (1987); B=Toluene/*n*C$_7$ (Toluene/*n*-Heptane); X=Xylene/*n*C$_8$; F=*n*-Heptane/MCH; H=Heptane.

ucts of the previous stage. In the early stages of the evaporative fractionation, the ratio of the low carbon number aromatics to the same carbon number *n*-paraffins (B and X in the Table 3) in the fractionated condensate, such as toluene/*n*-heptane, is less than that of the original crude oil without fractionation (Table 3 and Figure 6). With increasing fractionation, the carbon number of fractionated products is getting higher and higher. Among the fractionated products, the relative content of low-carbon *n*-alkanes is less and less, while the relative content of low-carbon aromatics is getting higher and higher, and the ratio of toluene/*n*-heptane in the fractionated products gradually increases. At the same time, the content of low-carbon number aromatics in the fractionation residue increases compared to the same carbon number *n*-alkanes. Therefore, the cross-plot of toluene/*n*-heptane to *n*-heptane/methylcyclohexane constructed by Thompson (1987) was essentially based on the stage products of the fractionation rather than the cumulative products of the fractionation.

As listed in Table 3, the toluene/*n*-heptane ratio of the fractionated product at different stages during the evaporative fractionation varies greatly. Crude oil with a low or very low toluene/*n*-heptane ratio does not mean that it is not a fractionated product, and a high toluene/*n*-heptane ratio could be indicative of a fractionation stage product, or it may be a residue of fractionation. In such a way, the Thompson's (1987) cross-plot will be problematic to determine whether the condensate is formed by evaporative fractionation, and also it cannot well identify whether the higher density crude oil is a stage product or a residual oil of evaporative fractionation, or a residual oil from which the liquid product that has been previously fractionated is re-fractionated by further gas-washing. Therefore, caution must be taken in terms of the application of the Thompson's (1987) cross-plot in geological conditions.

According to the principle of mass balance and the physicochemical properties of alkanes and aromatic hydrocarbons in the accumulative products during evaporative fractionation (e.g. polarity, fugacity, saturation vapor pressure), the toluene/*n*-heptane ratio should always be lower than that of the original crude oil, and the ratio in the residual oil should always be higher than that in the original crude oil. However, under geological conditions, there is a large uncertainty in the upward migration and re-accumulation of oil which experienced evaporative fractionation. If all or only the early fractionation part of oil and gas is accumulated, the toluene/*n*-heptane ratio of the condensate should be less than or equal to that of the original crude oil. If the accumulation is from one certain fractionation stage or the late stage of oil and gas, the toluene/*n*-heptane ratio of the condensate should be higher than that of the original crude oil. In most cases within geological frame only part of the fractionation of oil and gas can be re-accumulated, suggesting that re-accumulation in the
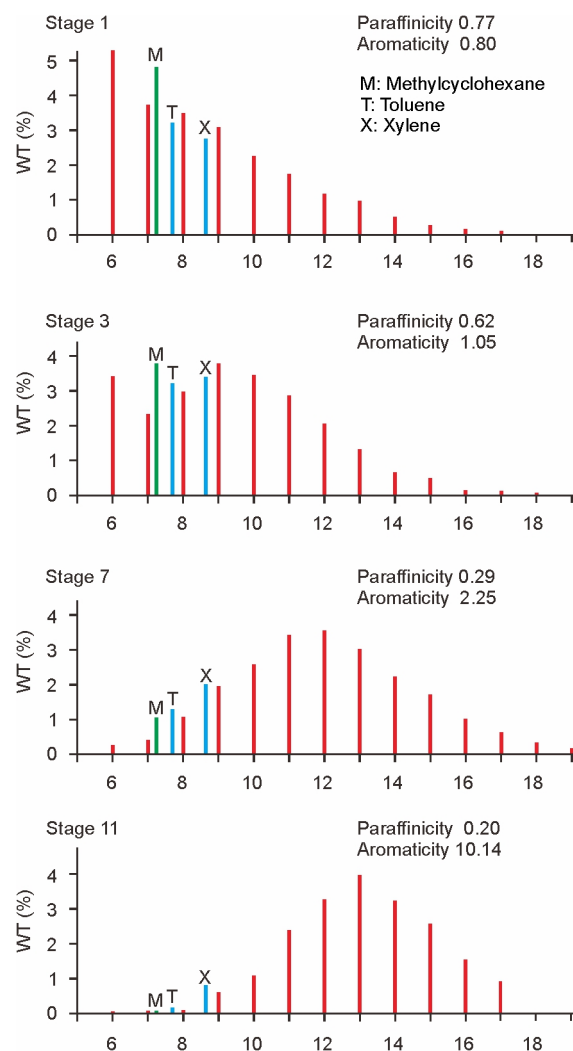

Figure 6 Simulated gas chromatograms of condensates from Experimental Series 3 (after Thompson, 1987).

trap is only one certain fractionation stage of oil and gas. Thereafter, the fraction of low-carbon aromatics in the condensate formed by evaporative fractionation is highly uncertain and its toluene/*n*-heptane ratio may be higher than, lower than, or equal to that of original crude oil. The intricate distributions of toluene/*n*-heptane ratios in condensates, light oils, waxy oils and heavy oils in Figure 5 well reflect the complexity of evaporative fractionation under geological conditions. A similar situation was observed in the South Marsh Island Block 208–239 area, offshore Louisiana, USA, showing that the toluene/*n*-heptane ratios of the condensate and crude oil were complicatedly distributed and many crude oils had toluene/*n*-heptane ratios greater than that of condensates (Losh et al., 2010).

In summary, to identify whether a condensate in one given petroliferous area is a product of evaporative fractionation cannot be simply based on its toluene/*n*-heptane and *n*-heptane/methylcyclohexane ratios in Thompson's (1987) crossplot. It is necessary to analyze the physicochemical properties

of oil and gas, the molecular distributions of different types of oil and gas, and the distributions of oil and gas with different physical and chemical properties in the studied region within actual geologic conditions.

5.1.4 The main formation mechanism of condensate, waxy and heavy oils

Although Thompson's (1987) cross-plot of the toluene/*n*-heptane vs. *n*-heptane/methylcyclohexane ratios is problematic to determine whether a condensate is formed by evaporation fractionation or not, if crude oil or condensate oil has unusually high or abnormally low content of low-carbon number aromatic hydrocarbons, regardless the influence of organic matter type and maturity, it still can be clear that the crude oil or condensate should experience a secondary alteration and evaporation fractionation could be the best candidate for this. Based on the review paper by Thompson (2010), the evaporative fractionations in many of the world's petroliferous basins were present and it is a relatively common secondary alteration of oil reservoir.

Many anticlinal structures in the central part of the southern margin of Junggar Basin contain normal crude oil, light oil, condensate, waxy oil and heavy oil. These crude oils have very similar carbon isotopic compositions and biomarker assemblages, indicating they are homogeneous mature oil derived from the Cretaceous lacustrine source rocks (Wang et al., 2013; Chen et al., 2015b, 2016a, 2016b, 2016c). Among these crude oils, the condensate oil is rich in low-carbon number *n*-alkanes and aromatic hydrocarbons. The high-density waxy oil and heavy oil relatively have less level of low-carbon number *n*-alkanes, but are relatively more enriched in low-carbon number aromatic hydrocarbons and high-carbon number *n*-alkanes (Figure 3 and Figure 4). These condensate oil, waxy oil and heavy oil have a higher toluene/*n*-heptane ratio (Figure 5), and are in agreement with the chemical composition of various liquid crude oils formed by evaporative fractionation.

On the other hand, large quantities of highly to over mature natural gases have been discovered in these anticlines. The natural gas was believed to be derived from the highly to over mature Jurassic coal-bearing source rocks, and the gas reservoirs are usually interbedded with various oil reservoirs, but not fully homogenized with crude oil (Chen et al., 2004; Liao et al., 2006, 2011; Wang et al., 2013). Therefore, it could be reasonable for the occurrence of evaporative fractionation in the central part of the southern margin of Junggar Basin due to the filling and accumulation history of liquid crude oil and natural gas.

Based on regional hydrocarbon generation history, it can be concluded that the oil reservoirs in the anticlines of the central part of the southern margin of Junggar Basin should be exposed to large quantities of natural gas. This means that the evaporative fractionation could be a main formation mecha-

nism of these condensates, while the high-density waxy oil is intermediates accompanying gas washing and the heavy oil are final residual crude oil in the reservoirs. The physical and chemical processes involved in the formation of condensates, waxy and heavy oils are summarized in Figure 7 and can be described as: The normal density crude oil reservoirs (A) is subject to large quantities of natural gas intrusion. Oil and gas are dissolved together to form light oil (B), and the deasphalting process may produce asphaltenes precipitation to form a small amount of heavy oil. The natural gas saturated with low molecular weight liquid hydrocarbons migrate upward, followed by the formation of condensates reservoirs in suitable traps (C1) as the temperature and pressure decrease, as exampled by the condensates reservoirs in the Hutubi and Manas anticlines. Finally, a natural gas reservoir (G) with little liquid hydrocarbons will be formed. The residual crude oil in the reservoir (B) forms high molecular weight *n*-alkanes-enriched waxy oil or heavy oil (C) due to the loss of light components. With the continuous injection of excessive natural gas and the enhancement of fractionation, the molecular weight of hydrocarbon carried by natural gas will become larger and larger. As the temperature and pressure decrease, waxy crude oil (C2) with little resins and asphaltenes can be formed under suitable conditions, typically as the waxy oils from the wells TG-1 and TG-2 in the Tugulu anticline and the Donggou Formation of Well H002 in the Horgos anticline (Table 1 and Figure 3). Similar waxy oil is also present in the offshore Louisiana (Losh et al., 2002, 2010). In addition, previous evaporative fractionation and re-accumulation of crude oil may also continue to be influenced by gas washings to form waxy oil with little resins and asphaltenes. Then, the natural gas saturated with liquid hydrocarbons continuously migrate upward to form condensate reservoirs (C3), and finally form natural gas reservoirs (G) that are substantially free of liquid hydrocarbons. At last, the crude oil in the original reservoir will form heavy oil (D) consisting mainly of high molecular weight *n*-alkanes, resins and asphaltenes, such as the heavy oil from the Ziniquanzi Formation in the Well H002 of Horgos anticline (Table 1 and Figure 3).

5.2 Phase-controlled fractionation

The "evaporative fractionation" proposed by Thompson (1987, 1988) is one of the formation mechanisms of the condensate oil with enrichment of low-carbon number *n*-alkanes, naphthenics and aromatics. This type of condensate can also be formed by another mechanism, phase-controlled fractionation (Larter et al., 1991). The term "phase-controlled fractionation" or "phase-controlled molecular fractionation" refers to the process in which the oil phase fluid containing gaseous hydrocarbons or gas phase fluid containing liquid hydrocarbons under high temperature and high pressure forms a gas saturated oil fluid and/or a condensate saturated
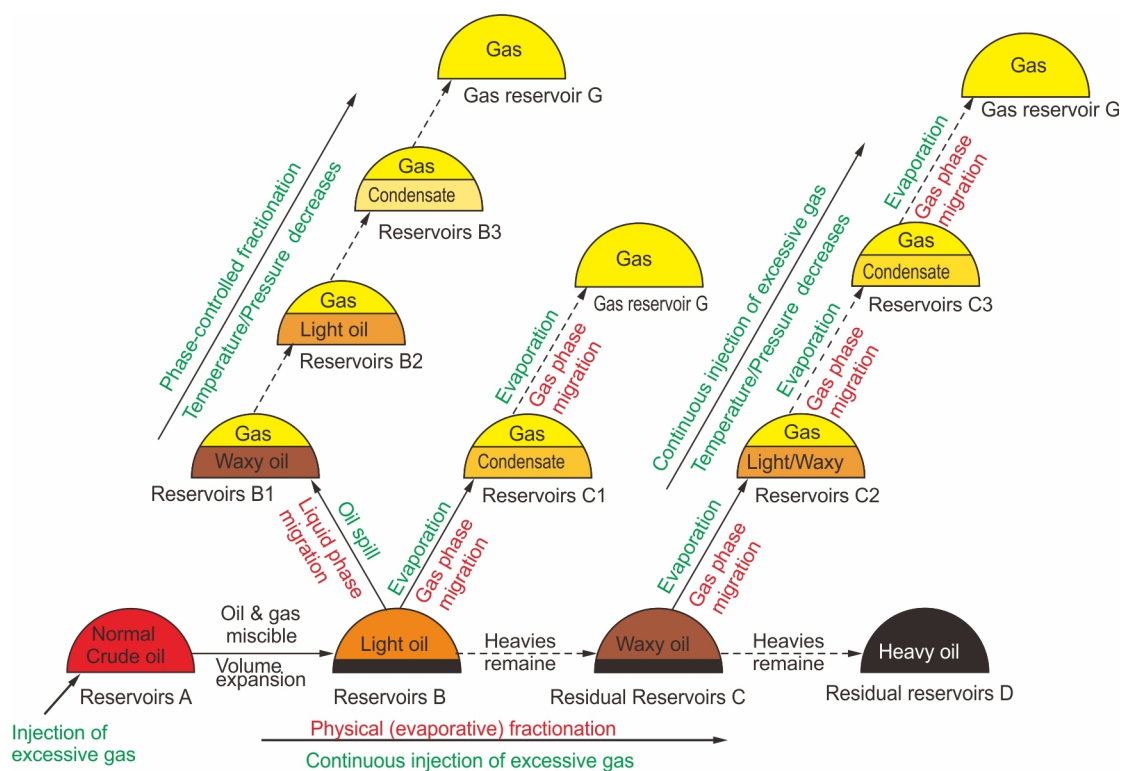
Figure 7   Schematic diagram of the formation process of condensate oil, light oil, waxy oil and heavy oil in the southern margin of Junggar Basin.

gas fluid when the dew point or bubble point temperature/pressure are reached. This is mainly induced by the decrease in the solubility of the minor phase in the major phase due to the decrease of temperature and/or pressure. Therefore, the migration media in phase-controlled fractionation can be either liquid phase or vapor phase. In fact, the "evaporative fractionation" proposed by Thompson (1987, 1988) is a special case of the phase-controlled fractionation, where the migration media is a vapor phase (natural gas) saturated by liquid hydrocarbons. In another word, once a large amount of natural gas intrusion into the oil reservoir, the low molecular weight liquid hydrocarbons are dissolved in the natural gas to form the vapor phase fluid containing liquid hydrocarbons, followed by upward migration and the liquid hydrocarbons are precipitated from the natural gas to form waxy oil, light oil, condensate and natural gas once reaching the dew point temperature/pressure. However, the migration media for the phase-controlled fractionation discussed later is liquid phase oil saturated by natural gas.

Larter et al. (1991) applied a high temperature and high pressure PVT apparatus to simulate the phase-controlled fractionation of a gas saturated liquid phase oil. The results showed that the light black oil was compressed with excess solution gas to form a gas-rich single-phase condensate, accompanied by asphaltenes precipitation. When petroleum migrated along the temperature and pressure drop of the "migration route" to a dew point where it reaches the two-phase equilibrium, partial liquid components was pre-

cipitated to form relatively higher density of crude oil, while the liquid component (condensate), which is still dissolved in the gas phase, is rich in low molecular weight aromatics. When the temperature and pressure further reduced, the natural gas and light crude oil which has less content of low molecular weight aromatics were formed. Su et al. (2004) conducted similar simulations and also obtained the low-molecular-weight aromatics-enriched condensate. Curiale and Bromley (1996a, 1996b) suggested that the condensate oils in the Vermilion 14 and Vermilion 39 blocks along the Louisiana coast of the Gulf of Mexico basin were the result of phase-controlled fractionation. Zhang et al. (2000) concluded that the condensate and waxy oils in the Tazhong and Tabei areas of Tarim Basin were also formed by evaporative fractionation/phase-controlled fractionation.

Part of the condensates and light crude oils in the central part of the southern margin of Junggar Basin may be directly formed by phase-controlled fractionation (Figure 7). The normal oil reservoir (A) is subject to a large amount of natural gas intrusion, which produces an oil and gas mixture, and the crude oil becomes lighter in quality, resulting in the formation of a light oil-dominated reservoir (B). On the other hand, the deasphalting in the gas invasion process leads to the formation of heavy oil (residual oil) in the reservoir. At the same time, the gas intrusion induces the expansion of the crude oil volume, suggesting that part of the gas saturated light oil can spill out of the reservoir and migrate upwards as liquid phase along the carrier beds, faults or fractures. With

the decrease in temperature and pressure, the liquid compositions with larger molecular weight are gradually precipitated to form waxy crude oil (Reservoir B1). The low and medium molecular weight liquid constituents continue to migrate as liquid phase and are precipitated at lower temperatures and pressures to form light crude oil (Reservoir B2) or condensate oil (Reservoir B3). Eventually, it will be possible to form a natural gas reservoir (G) with little liquid hydrocarbons.

In the central part of the southern margin of Junggar Basin, it can be found that the "phase-controlled fractionation" could account for mechanism on formation of various types of crude oils and condensate in several cases. For example, the crude oil in the 3064–3067 m interval of Well H10 has a density of only 0.7967 g/cm³, which is almost the same as that of the condensate (Table 1), but its color is brown-black, significantly deeper than the general condensate. The contents of resins and asphaltenes in this oil are very low, only slightly higher than that of the condensate, but the relative content of high-carbon number *n*-alkanes in this oil is higher than that in condensate (Figure 3), as well as a relatively high content of low-carbon number aromatic hydrocarbons (Table 2). If this kind of light crude oil is further subjected to gas invasion, evaporative fractionation may result in the formation of waxy oil with a little resins and asphaltenes. For example, the crude oils from the wells TG-1 and TG-2 in the Tugulu anticline and the Cretaceous Donggou Formation of the Well H002 in the Horgos anticline (Table 1), contain very low content of resins and asphaltenes, and are almost depleted of low-carbon number *n*-alkanes, however, they are rich in low-carbon number aromatics (Figure 3). They may be waxy oil formed by the phase-controlled fractionation from normal crude oil, while the heavy oil in the Well H002 may be formed by deasphalting.

6. Geological conditions and processes of condensates formation

In the southern margin of Junggar Basin, the source rocks of the Permian, Triassic, Middle-Lower Jurassic, Qingshuihe formation of the Lower Cretaceous and the Anjihaihe Formation of the Paleogene are all effective hydrocarbon source rocks. The Permian, Triassic and Jurassic source rocks in the depression are deeply buried and have reached highly to over mature stages (Wang et al., 2013; Chen et al., 2015a, 2016c). The maturity of Cretaceous source rocks is relatively low, at present mainly in mature oil generation stage. The Paleogene source rocks have lower maturity and are mainly in the immature-low mature stage except a few in mature stage. The correlation between hydrocarbon generation history of these source rocks and tectonic/structure evolution determines whether the generated oil and gas can be accumulated in the structures, then the origin and formation process of condensate oil, waxy oil and heavy oil.

6.1 Hydrocarbon generation history of the source rocks

In the southern Junggar Basin, the paleogeothermal field experienced a gradual descending process from high to low, and the geothermal gradient gradually decreased from 32.0–36.6°C/km in the Permian to 18–22°C/km at present (Wang et al., 2000a, 2000b; Qiu et al., 2001, 2002). In order to determine the thermal evolution and hydrocarbon generation processes of source rocks in the southern depression, hydrocarbon generation history in the southern depression was calculated by using the basin modeling under the constraints of the vitrinite reflectance of the well samples (Figure 8). In the south of the Manas anticline, the oil window of the Middle and Lower Jurassic source rock is manly in the Cretaceous-Miocene (130–10 Ma), and the peak of oil generation is in the Eocene-Early Miocene (50–15 Ma). The natural gas generation by thermal degradation of kerogen occurred at the beginning of the Eocene (50 Ma), and is now in the stage of maximum generation of natural gas (Figure 8a). Due to the deep-burial of the Permian and Triassic source rocks compared to that of the Jurassic source rocks, their oil and gas generation window is earlier than that of the Jurassic one. The maximum burial depth of the Lower Cretaceous source rocks reaches 8800–9000 m, which is about 800–1000 m deeper than that in the Changji sag. The source rocks began to generate oil in the Eocene (50 Ma), and entered peak oil at the beginning of the Miocene (20 Ma). At present, the Cretaceous source rocks are mainly in the peak of oil generation and partly in the late stage of oil window. The source rocks of the Paleogene Anjihaihe Formation began to enter the oil generation threshold at the end of the Miocene (6 Ma), and are still in the early stage of oil generation. The depth of hydrocarbon source rocks in the northern part of the depression is shallower than that in the south of the Manas anticline and the hydrocarbon generation time is slightly later (Figure 8b). The oil generation time of the Lower-Middle Jurassic source rocks is also mainly before the Pliocene, and the Cretaceous is in the low mature stage.

6.2 Tectonic evolution and hydrocarbon accumulation

The northern Tianshan-Bogda Mountains in the southern margin of the Junggar Basin began to rise significantly since the Miocene (25 Ma) (Hendrix et al., 1994; Fang et al., 2005, 2007; Hu et al., 2005; Guo et al., 2006, 2011; Du et al., 2007; Wang et al., 2008). The first row of structures such as Qigu anticline was formed at about 10–7 Ma; and the second row of structures such as the Horgos anticline was formed at about 3–1.5 Ma. The Dushanzi and Anjihai anticlines are the third row of structures which were formed at about 1 Ma, and the Hutubi anticline was formed later. Therefore, the anti-
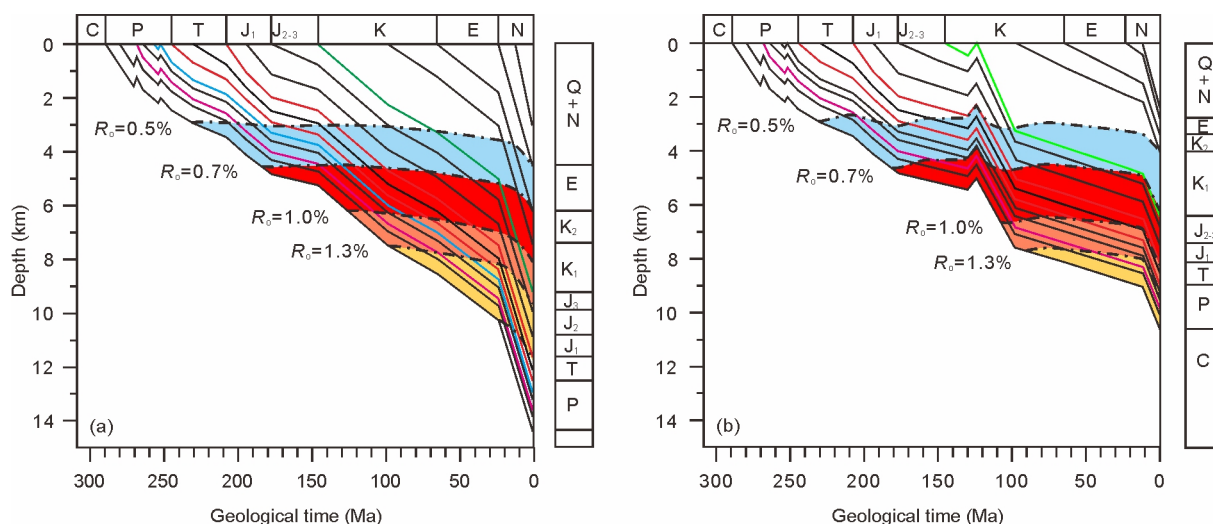
Figure 8   Burial history and thermal evolution of the source rock in the central part of the southern margin of Junggar Basin. (a) the south of the Manas anticline; and (b) the northern part of the Southern Depression.

clines in the southern margin of Junggar Basin were formed very late and can capture only oil and gas generated since 10 Ma.

It is clear that, during the main oil generation stage of the Permian, Triassic and Jurassic source rocks, all anticlinal structures have not been formed, excerpt for the anticlines where the Permian and Triassic were buried relatively shallow with occurrence of early ancient anticlines. This means that most of anticline structures are basically impossible to accept crude oils derived from the Permian, Triassic and Jurassic source rocks. However, the main oil generation stage of the Cretaceous source rocks matches well with the formation period of these structures, suggesting that these structural traps can accumulate the crude oils derived from Cretaceous source rocks. Oil-source correlation confirmed that mature oils in these traps were derived from lacustrine source rocks of the Cretaceous Qingshuihe Formation (Wang et al., 2013; Chen et al., 2016a, 2016b, 2016c).

On the other hand, Jurassic coal-bearing source rocks have been in highly to over mature stage since the Pliocene, and they also match well with the formation period of anticline structures in the southern margin of Junggar Basin. These anticlines can capture natural gas derived from the Jurassic coal-bearing source rocks. Natural gas in these anticlines contains 80–97% of methane, and is dominated by wet gas. The $\delta^{13}C$ values of the methane in natural gas are mainly between −35‰ and −31‰, and the $\delta^{13}C$ values of ethane and propane are greater than −25‰, indicating that they are typically humic-type gas and were derived from Jurassic coal-bearing source rocks (Wang et al., 2004, 2013; Chen et al., 2004; Liao et al., 2006, 2011).

In addition, the faults in the southern margin of Junggar Basin were well developed, and each anticline structure has a series of thrust faults and secondary faults parallel to the anticline axis (Figure 3), connecting deep hydrocarbon source kitchens to upper reservoirs. These faults are favorable channels for upward migration of oil and gas, and provide good conditions for oil and gas reservoirs to be multiple-charged. Using fluid inclusions and fluorescence spectral characteristics of the sandstone reservoir of the Manas anticline oil and gas reservoirs, Bai et al. (2013) concluded that there were two periods of oil and gas charging in the reservoirs from the Manas anticline. The first period (11 Ma) was the charge of crude oil, and the second period (3 Ma) was the filling of natural gas. Because other anticlines in the central part of the southern margin of Junggar Basin have almost the same tectonic background and history as the Manas anticline, they should have similar process of hydrocarbon accumulation. Although the structure formation time and the hydrocarbon filling time may not be very accurately determined, the oil and gas inclusions revealed that the central anticlinal structures of the southern margin of Junggar Basin had undergone two processes of charging and accumulation, namely, the first oil and the later gas (Bai et al., 2013).

6.3   Geological process of condensate formation

The evaporative fractionation/phase-controlled fractionation is an extremely complex physical and chemical process (Figure 7). The natural gas injected into the oil reservoir is different in its dryness or composition, and its ability to carry liquid hydrocarbons is also different. The amount and duration of natural gas injection varies considerably, and the composition of the liquid hydrocarbons carried out and the residues in the reservoir vary considerably. Liquid hydrocarbons carried by natural gas can form crude oils with different chemical composition in traps at different temperature/pressure regime, which may be condensate oil, light oil or waxy oil. If these re-pooled crude oil continues or is re-subject to natural gas washing, its composition will make further change, and it

may form waxy oil dominated by high carbon number *n*-alkanes, light oil or condensates. Therefore, the evaporative fractionation/phase-controlled fractionation can lead to a complex situation of the mixed distribution of various types of oils in one given region.

The anticlinal structures in the central part of the southern margin of Junggar Basin have perfect geological and geochemical conditions for the formation of condensate oil, light oil, waxy crude oil and heavy oil by evaporative fractionation or phase-controlled fractionation. The oil and gas filling and accumulation may have undergone the following processes: Firstly, the Cretaceous lacustrine source rocks in the Miocene began to generate large amounts of crude oil, and these crude oils fill into the reservoirs in the anticlinal traps to form oil reservoirs. Secondly, the Jurassic coal-bearing source rocks in the Pliocene began to generate a large number of high-mature natural gas, and then the gas migrates upward along the faults and fractures to fill in the early reservoirs with Cretaceous crude oil, resulting in a gas invasion into the reservoirs for the occurrence of the evaporative fractionation/phase-controlled fractionation. Thirdly, the mixed oil and gas in gas-phase or liquid-phase continues upward migration, and with decrease in temperature/pressure, precipitation of the liquid compositions directly leads to the formation of waxy oil, light oil and condensates. As a result, this process results in the co-occurred complexity of normal crude oil, waxy oil, light oil, condensate oil, heavy oil and natural gas, with staggered distributions in many anticlinal structures in the southern margin of Junggar Basin.

7.   Conclusions

The anticlinal structure traps in the southern margin of the Junggar Basin are well developed. There are normal crude oil, light oil, condensate oil, waxy oil, heavy oil and large amounts of natural gas in many anticlines in the central part of the southern margin of Junggar Basin. Based on the study of detailed geochemical characteristics of more than 40 various oils, the following conclusions can be drawn as to the origin and formation mechanism of condensate, waxy oil and heavy oil in this area.

(1) The condensates are dominated by low carbon number hydrocarbons, but the alkanes above C_{15} are also relatively high. The C_{29}-steranes 20S/(20S+20R) value is between 0.40 and 0.55; the maturity of equivalent vitrinite reflectance (R_c) calculated by the aromatics MPI and MPDF is 0.70–1.1%; the heptane and the isoheptane values range from 19% to 21% and from 1.9 to 2.1, respectively; and the formation temperature of the condensate oils is between 120°C and 130°C. All above indicate the condensate oils are mature rather than highly mature.

(2) The condensates are rich in naphthenics and toluene,

xylene and other low-carbon aromatics. Their toluene/*n*-heptane ratios are between 1.5 and 2.0, and the distribution of alkanes is mirror image with high density waxy oil and heavy oil, suggesting that they are formed by the evaporative fractionation/phase-controlled fractionation of the early accumulated crude oil. The waxy oil is an intermediate or stage product of the fractionation process, and the heavy oil is the final residue of the fractionations. Evaporative fractionation is the main formation mechanism of these various oils, and phase-controlled fractionation is the secondary formation mechanism.

(3) In the southern margin of Junggar Basin, the periods of oil generation of the Cretaceous lacustrine source rocks and gas generation of Jurassic coal-bearing source rocks match well with the formation period of anticline structures. Cretaceous crude oils previously accumulated experienced large quantities of natural gas invasion from highly to over mature Jurassic coal measures. The evaporative/phase-controlled fractionation results in the complicated situation of coexisting and staggered distribution of various types of crude oil and natural gas in the anticlines.

(4) Evaporative/phase-controlled fractionation can lead to significant changes in the content of light hydrocarbon compounds such as heptane, methylcyclohexane, and toluene, etc. The toluene/*n*-heptane ratio of the precipitated oil is highly uncertain, depending on the stage of fractionation in which the oil is trapped. The origin of the condensate oil cannot be simply determined by its ratios of toluene/*n*-heptane and *n*-heptane/methylcyclohexane on Thompson's (1987) crossplot, but it can be comprehensively determined by the aspects of geological background, thermal history of source rocks and petroleum generation, physical and chemical features, and vertical and lateral distribution of various crude oils in the area.

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