Multistage petroleum charges in the Silurian of Tazhong north slope of the Tarim basin, northwest China: evidence from fluid inclusions and organic geochemistry

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Abstract Well S provides a good opportunity to investigate the petroleum filling history in Tazhong North Slope. Petrographic investigations suggest that bitumen, oil, and petroleum inclusions coexist in Silurian sandstones from well S, reflecting a complicated reservoir filling history in the study area. Integration of organic geochemistry and fluid inclusion analysis shows that the Silurian reservoir has experienced three episodes of petroleum charge, that is from the late Silurian to the early Devonian, the early-middle Triassic and the Paleogene, respectively. The present-day reservoir fluids in the Silurian are the mixtures generated in multiple (at least two) episodes of petroleum charge. The oil charging into Silurian reservoir in the early period had experienced considerable degradation, and was mixed with later non-degraded oil.

Key words Fluid inclusion, Organic geochemistry, Silurian, Tazhong North Slope, Tarim Basin

1 Introduction

Tazhong area, which consists of Tazhong Uplift and its north slope, is one of the most important petroleum pay regions in Tarim basin; many oil wells have been discovered there (Fig. 1). However, because of complex geological background, oil & gas pools in this area often show a complicated reservoir filling history (Xiao et al., 1996; Jia et al.,1997). The Tazhong Uplift can accommodate oil not only from Cambrian-lower Ordovician and middle-upper Ordovician source rocks of itself, but also from the adjacent depressions (e.g. Manjiuer Depression). Furthermore, as some of the faults cut through all the lower Palaeozoic strata, the oil in any given reservoir rock may be actually a mixture of oils from different sources and/or episodes, forming a hybrid petroleum system (Xiao et al.,2000).

The Silurian reservoir is most close to the Cambrian-lower Ordovician and middle-upper Ordovician, which are the two sets of most important source rocks of the Tarim basin. As a result, every petroleum event related to hydrocarbon generation and expulsion must be preserved as fossil records (bitumen or petroleum inclusions) in Silurian reservoir rocks. Previous studies have indicated that the bitumen and oil in the Silurian

Fig. 1 Sketch map showing the structural outline of Tazhong area and the location of well S [modified after Xiao et al (2000) and Cai et al(2005) ]
reservoir are of lower Palaeozoic sources (Liu, 1998). So reconstruction of Silurian reservoir filling history is of vital significance in understanding the forming mechanism of oil & gas pools in the Tazhong area.

Well S is an important exploratory well of SINOPEC and is located at Tazhong North Slope (Fig. 1), with Devonian and Silurian strata as the chief target. In this completed well, a small amount of heavy oil was acquired, and various levels of oil shows can be detected actively in the lower sand member of the Silurian (Chen et al., 2005). So the well provides a good opportunity to investigate petroleum filling history in Tazhong North Slope. In this paper, a combination of fluid inclusions and organic geochemistry analyses of the materials from this well are used to illuminate the characteristics of multiple petroleum charges in the Silurian reservoir.

2 Samples and methodology

2.1 Samples

The studied sandstone samples belong to the middle Silurian Yinmangdian Formation (S2y) and the lower Silurian Titaeritage Formation (S1t). Eight samples were cored at the depths between 4960.13 and 5336.49m from well S. Fundamental data for these samples are listed in Table 1.

<table>
<thead>
<tr>
<th>No.</th>
<th>Age (m)</th>
<th>Lithology</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4960.13</td>
<td>greyish-white medium grain quartzite sandstone</td>
</tr>
<tr>
<td>2</td>
<td>4961.13</td>
<td>red medium grain sandstone</td>
</tr>
<tr>
<td>3</td>
<td>5047.26</td>
<td>red medium-fine grain sandstone</td>
</tr>
<tr>
<td>4</td>
<td>5234.24</td>
<td>greyish-white argillaceous medium-coarse grain quartzite sandstone</td>
</tr>
<tr>
<td>5</td>
<td>5319.51</td>
<td>greyish-white medium-coarse grain quartzite sandstone</td>
</tr>
<tr>
<td>6</td>
<td>5329.37</td>
<td>greyish-white oil-bearing medium-coarse grain sandstone</td>
</tr>
<tr>
<td>7</td>
<td>5336.36</td>
<td>greyish-white oil-bearing medium-coarse grain sandstone</td>
</tr>
<tr>
<td>8</td>
<td>5336.49</td>
<td>greyish-white oil-bearing medium-coarse grain sandstone</td>
</tr>
</tbody>
</table>

2.2 Analytical methods

2.2.1 Organic geochemistry

Reservoir sandstones were crushed into small pieces with the size of ca. 100 mesh (approximately 0.172mm), and organic materials were extracted from them by chloroform for 48h. The extracts were first precipitated in petroleum ether to eliminate the asphaltine, and then fractionated by column chromatography using alumina oxide/silica gel. A sequential purge by petroleum ether, dichloromethane, and chloroform was conducted so as to separate saturate and aromatic hydrocarbons and NSO fractions, respectively. Saturate fractions were analyzed on a Finnigan MATSSQ 710 gas chromatograph-mass spectrometer (GC-MS). We used a DB5 fused silica column (30m×0.32mm i. d.) coated with a 0.25μm-thick film. The temperature program was set as follows: first, the temperature was held for 1 min at 100°C, then increased at a rate of 3°C/min to 220°C, followed by 2°C/min from 220°C to 300°C and kept constant for 20min. The injector temperature was 300°C, and the carrying gas was helium.

2.2.2 Fluid inclusion measurement

Doubly-polished wafers with a thickness of ca. 70 - 100μm were prepared from core samples from well S and were examined using a Leica petrographic microscope equipped with both transmitted light and ultraviolet (UV) light. UV light for fluorescence analysis was excited by a mercury lamp, and filters were used so that only long wavelength UV light (transmission maximum at 366nm) was admitted to reach the samples.

Microthermometric measurements were acquired with a Linkam THMS600 (Linkam, UK) heating/cooling stage attached to a conventional optical microscope. The stage was calibrated by synthetic pure H2O inclusion. Ice-melting and homogenization temperatures published in this paper are accurate to ±0.2°C.

3 Present-day reservoir fluids

Petrographic observation of S1t sandstone samples indicates yellow-brown oil and black bitumen were abundant in the pores which were not cemented by calcite. The oil fluoresce intense orange color under UV light (Fig. 2). In pores where two kinds of hydrocarbons coexist, the bitumen occupies the main space, while the oil occurs around the bitumen or within the microfractures inside quartz grains. The hydrocarbons and its distribution suggest that the Silurian reservoir experienced at least two episodes of petroleum charge, which were characterized by bitumen for the early episode and by oil for the later one.

Organic geochemistry data also imply that the Silurian reservoir of well S experienced multiple petroleum charges. From the total ionization chromatogram (TIC) of the saturate fraction of core extracts from S1t sandstones, we can see that the chromatogram baseline drifts upward, showing an obvious hump (unsolved complex mixture reflecting degradation) (Fig. 3a).

However, the hump was superimposed by a well-defined suite of n-alkanes, as shown clearly in the m/z 285 mass chromatogram (MS) (Fig. 3b). According to Chen et al. (2005), the chromatogram of oil samples from the same reservoir sandstone also exhibits a similar shape. These features suggest that the present-day reservoir fluids in the Silurian belong to a mixture of a degraded and a fresh oil, represented by the complex mixture and the well-defined n-alkanes, respectively (Zhang et al., 2003; Parnell et al., 2005). Organic geochemistry analysis indicates that the Silurian reservoir had experienced multiple (at least two) episodes of petroleum charges. The oil charged into the Silurian reservoir earlier had experienced considerable degradation, and was mixed by fresh oil in the later episode (s).

4 Fluid inclusion analysis

4.1 Diagenesis

Understanding of diagenesis is an important step in fluid inclusion analysis. Especially in petroleum environments, diagenesis is the essential criterion to classify the generations of petroleum inclusions. The diagenetic features of Silurian sandstones of Tazhong area have previously been described by
Fig. 2  Microscopic characteristics of reservoir fluids

Fig. 3  Total ionization chromatogram (a) and m/z85 mass chromatogram (b) of core extracts from S_1t sandstones. Pr- pristane, Ph- phytane

Cai, et al. (1999) and Zhang, et al (2005). Fig. 4 shows the diagenetic sequence of Zhang et al. (2005). Shortly after the deposition of the Silurian, clay rings have appeared around most skeletal detritals, followed by the precipitation of early calcite cements. The consequence of mechanical compaction to detrital grains is not only directional arrangement and pressure solution, but also their distortion and fracturing. After the solution, a pore-filling chlorite precipitated, and chloritization of biotite may occur during this period. The late diagenesis is characterized by the precipitation of quartz overgrowth, ferrocalcite and ankerite filling the residual pores. And a large amount of illite appeared in the late diagenesis.

Diagenetic minerals in samples of well S consist successively of clay rings around quartz grains, early calcite cements, quartz overgrowth, ferrocalcite and ankerite. Quartz overgrowth and ferrocalcite are extremely common, and can be observed in all samples. Early calcite cements are localised in all of the S_1t samples. And ankerite is only found in the 5234.24m sample.

4.2 Description of fluid inclusions
Petrographic examination of the Silurian samples reveals the presence of two distinct types of fluid inclusions-aqueous and petroleum. At room temperature, most aqueous inclusions contain an aqueous liquid and a vapour bubble that occupies <5 volume percent of the inclusion. The inclusions of this type are mainly used for microthermometric analysis, and will not be described in detail.

Based on the occurrence and optical properties, the petroleum inclusions can be subdivided into four types.

Type I petroleum inclusions are mainly localised within healed microfractures inside detrital quartz grains, or are clustered in the early calcite cements. The abundance of type I petroleum inclusions is low, and the GOI (grains containing oil inclusions) value is below 0.5% in S_2y samples. However, in S_1t samples the GOI value can be as high as 20 ~ 30%. Type I inclusions all belong to dark-brown liquid hydrocarbon inclusions (Fig. 5A), and display no fluorescence under UV light.

Type II petroleum inclusions are localised within healed quartz microfractures that crosscutting the overgrowths. They contain a liquid hydrocarbon, or a hydrocarbon liquid and a gaseous bubble. They are pale-yellow or clear under transmitted light, and fluoresce bright pale-yellow or white-blue colour when excited by UV light (Fig. 5B-D). Nevertheless, Type II inclusions are rare in all samples, and the GOI value is approximately 0.5%.
Type III petroleum inclusions are distributed abundantly (GOI value 8 ~ 10%) in healed quartz microfractures, quartz overgrowth, early calcite cements and late ferroan calcite. This type of inclusions contain a hydrocarbon liquid and a gaseous bubble at room temperature (Fig. 5E-G). They can be readily identified as petroleum owing to their specific optical properties. Even under transmitted light, they are brown yellow or dark brown. Under UV light, a faint dark-brown fluorescence can be observed.

Type IV petroleum inclusions are occasionally seen in the ferrocalcite. They are grey gaseous inclusions with extremely low GOI value (Fig. 5H).

4.3 Microthermometry of aqueous inclusions

Microthermometry was conducted on aqueous inclusions which coexist with petroleum inclusions in early calcite cements, quartz microfractures and late ferroan calcite, respectively. And the results are shown in Fig. 6. All analysed inclusions are two-phase ones at room temperature, and the homogenization always occurs to liquid phase. Due to the similar shape and similar microthermometry results, the results for aqueous inclusions in the early calcite and those in the late ferroan calcite were put together in the histogram. As shown in Fig. 6, the aqueous inclusions in calcites were homogenized to liquid phase over the range of 90 ~ 110°C, with the majority between 90 and 100°C and vary in salinity from 14.5 to 21 wt% NaCl equivalent. The homogenization temperature of aqueous inclusions in quartz microfractures shows a bimodal distribution, with the peaks around 100 ~ 110°C (A) and 120 ~ 130°C (B). On the homogenization temperature vs. salinity diagram we can see two groups of inclusions; the former is characterized of 95 ~ 120°C and 13 ~ 20 wt% NaCl (A) whereas the latter 115 ~ 135°C and 3 ~ 10 wt% NaCl (B). The group A inclusions occur in quartz microfractures that do not cut through the rims of detrital quartz whereas group B inclusions crosscut the quartz overgrowths.
4.4 Discussion of multiple petroleum events

Petroleum inclusions are the direct evidence for migration of petroleum fluids through the strata in the geological past (Melimans, 1987; Bodnar et al., 1990; Pan et al., 2006). The four types of petroleum inclusions with distinct features in the Silurian samples have faithfully recorded multistage petroleum activities in Tazhong North Slope. The distribution patterns of microthermometric data reflect different temperature and salinity conditions of paleofluids related to petroleum events.

Fluid inclusion microthermometric data are often used in combination with burial and thermal history of reservoir to estimate the timing of petroleum charge (e.g., Nedkvitne et al., 1993; Cao et al., 2005, 2006). In general, the homogenization temperature only represents the minimum trapping temperature of a fluid inclusion. However, in petroleum provinces, as the aqueous fluid phase is often saturated with gas [commonly methane, Hanor (1980)], the homogenization temperature of aqueous inclusions may reasonably be taken as the trapping temperature (Nedkvitne et al., 1993; Pirronon, 2004) and pressure correction is not necessary. This assumption is made for well S. According to the distribution patterns of microthermometric data mentioned above (i.e., the homogenization temperatures around 90 ~ 110°C and those around 120 ~ 130°C for two populations of aqueous inclusions) and combined with the burial and thermal history simulated by the “BasinMod ID” software, it is suggested that there were two episodes of petroleum charge occurring during the early-middle Triassic and the Paleogene (Fig. 7); the first episode is also indicated by type I petroleum inclusions whereas the second episode by type II & III inclusions. Due to the low GOI value, type IV gaseous inclusions may be attributed to an oil-cracking process (Zhang et al., 2004; Xiao et al., 2005a; Zhao et al., 2006).

More and more researches indicate that an earlier petroleum event occurred in Tazhong area (e.g., Jia et al., 1997; Xiao et al., 1997, 2005b; Xiao et al., 2000; Liu et al., 2000). The oil and gas from Cambrian-lower Ordovician source rocks migrated into lower Silurian sandstones to form petroleum reservoirs during the late Silurian to the early Devonian. But during the early Hercynian movement, strong uplift occurred in this area caused erosion of cap rocks; as a result, the reservoirs were breached and some of them were even destroyed. The wide occurrence of residual bitumen-bearing sandstones of the early Silurian age is the evidence for the existence of palaeo-oil and gas pools. And this is the case in well S. As we can see in Fig. 2, the bitumen is mainly distributed in primary pores of Silurian sandstones, which is obviously correlated to the earliest petroleum event. But fluid inclusions were not entrapped during this petroleum event.
owing to shallow burial and weak diagenesis of Silurian sandstones at that time (Xiao et al., 1997; Liu et al., 2000).

Thus, the Silurian reservoir of well S actually experienced three episodes of petroleum charge, i.e., the late Silurian to the early Devonian, the early-middle Triassic and the Paleogene.

5 Conclusion

(1) Well S provides a good opportunity to investigate petroleum filling history in Tazhong North Slope. Petrographic investigations suggest that bitumen, oil, and petroleum inclusions coexist in Silurian sandstones from well S, reflecting a complicated reservoir filling history in the study area.

(2) Organic geochemistry reveals that the present-day reservoir fluids in the Silurian are the mixtures of petroleum charge in multiple episodes. The oil charging into Silurian reservoir earlier had experienced considerable degradation, and was mixed by later charged non-degraded oil.

(3) Based on the fluid inclusion analysis, the Silurian reservoir of well S experienced three episodes of petroleum charge with the timing of from late Silurian to early Devonian, the early-middle Triassic and the Paleogene, respectively.

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