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# Formation and evolution of Silurian paleo-oil pools in the Tarim Basin, NW China $^{\bigstar}$

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ARTICLE INFO

Article history: Received 13 December 2006 Received in revised form 27 May 2008 Accepted 30 May 2008 Available online 10 June 2008

#### ABSTRACT

The formation and evolution of Silurian sandstone reservoirs, which are characterized by the coexistence of reservoir bitumen with different types of commercial oil pools in a very extensive area around the slope and uplift areas of the Manjiaer Depression in the Tarim Basin, NW China, are still in question. In this paper we report our investigations on the geological and geochemical characteristics of this reservoir. The results show there are four types of oil shows in the reservoir: severely biodegraded solid bitumen, biodegraded heavy oil, normal oils and light oils with slight biodegradation. Three suites of petroleum inclusions were identified in the reservoir which were formed during the early Devonian, Triassic to Cretaceous and late Tertiary, respectively. Petroleum in the Silurian sandstone reservoirs mainly originated from the mid-lower Cambrian source rock and mid-upper Ordovician source rock. The thermal maturity evolution of the source rocks in slope areas has shown that there were three main stages of oil generation that controlled the formation of the above three stages of petroleum inclusions. This evidence indicates that the Silurian reservoir has experienced three petroleum charge events during its geological history. The first charge event occurred during the early Devonian and it resulted in giant paleopools, which gradually evolved into the presently widespread bitumen due to the combination of biodegradation and thermal alteration. The second charge covered a long period from Triassic to Cretaceous and probably was related to both the mid-lower Cambrian source rock and the mid-upper Ordovician source rock. The petroleum generated during this stage mixed and interacted with the low matured residual bitumen and eventually formed the present soft bitumen and some residual heavy oils in the reservoirs. The third charge event, mainly associated with the mid-upper Ordovician source rocks, occurred mainly during late Tertiary and accounted for most of the present commercial petroleum pools (mainly normal and light oils).

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# 1. Introduction

The Tarim Basin, NW China, is a large lapped basin with a total thickness of 16 km of Paleozoic, Mesozoic and Cenozoic sediments. It has experienced multiple uplift and subsidence episodes that resulted in several sets of source-reservoir-cap assemblages and multiple petroleum generation-migration-accumulation events (Kang and Kang, 1996; Li et al., 1996). Commercial oil/gas pools have been discovered in strata from Cambrian to Tertiary (Jia, 1999). Among them, the Silurian sandstone reservoir is quite unique because it contains abundant solid bitumen (called bituminous or asphaltic sandstone by Chinese geologists). Following the first discovery of the bituminous

<sup>\*</sup> An earlier version was presented at the AAAPG in Beijing, 2004.

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<sup>0146-6380/\$ -</sup> see front matter © 2008 Elsevier Ltd. All rights reserved. doi:10.1016/j.orggeochem.2008.05.011

sandstones from the well Ha1 in the Tabei Uplift in 1991, this type of reservoir has been discovered in other structural units, such as the Tazhong, Mandong and Lunnan areas (Guo et al., 2001; Liu et al., 2001). It was believed that the bitumen originated from paleo-pools which were widely distributed in the slope and uplift areas around the Manjiaer Depression and had been altered by biodegradation during late Devonian (Liu et al., 2001).

Since the occurrence of solid reservoir bitumen represents a major exploration risk in many hydrocarbon reservoirs (Lomando, 1992), it seems hopeless to have commercial oil potential for the Silurian reservoirs. However, some commercial oil pools, containing normal, heavy, and light oils, have been discovered in this reservoir from the northern slope area of the Tazhong Low Uplift (Zhang et al., 2002). More recently gases cracked from oil associated with Cambrian source rock have been reported in well MD1 in Silurian reservoirs of the eastern Manjiaer Depression (Xiao et al., 2005). Earlier oil source correlations based on biomarker compounds showed that all the produced marine oils, including the extracts of Silurian bituminous sandstones, originated from the mid-upper Ordovician source rocks (Zhang et al., 2000a). The mid-upper source rocks developed mainly on the slopes of paleo-uplifts with current burial depth of 4500-5500 m and their maturities are mostly in the oil window (Zhang et al., 2004). Therefore, it is unreasonable for them to source the Silurian reservoirs before late Devonian. This implies that there are probably multiple oil charge events, associated with higher maturity source rocks of both Cambrian and mid-upper Ordovician ages, to form the complicated oil shows in the reservoir. In fact several questions remain to be fully addressed including when the paleo-oil pools and the present commercial oil pools formed, how the heavy oil was formed, and what are the relationships between the bitumen and the later charged petroleum fluids. These questions are not only of academic significance, but they are also related directly to the evaluation of the commercial potential of the Silurian reservoirs in the Tarim Basin.

In the present paper, petrological and geochemical methods were combined to investigate the multiple events of petroleum charge history of the Silurian reservoir and their contribution to the formation of current oil pools. Finally, a geological and geochemical model was suggested for the formation and evolution of the paleo-oil pools in Silurian reservoirs.

# 2. Geological setting

The study areas are focused around Manjiaer Depression (Fig. 1A), including the Tazhong Low Uplift (Tazhong area), the Lunnan Low Uplift (Lunnan area), the northwest slope of the Manjiaer Depression (Manxi area), the eastern part of Manjiaer Depression (Mandong area) and the Yingjisu Depression. These structural units have different structural-sedimentary histories.

The Manjiaer Depression is largely characterized by continuous subsidence and it experienced a complete marine transgression-regression depositional cycle with nearly 16 km thickness of sediments from Sinian (Precambrian, 542–680 Ma) to Quaternary (Kang and Kang, 1996). The Silurian strata, about 1000 m in thickness, developed with unconformable boundaries both at bottom and top (Fig. 1B). The Yingjisu area was the eastern slope of the Manjiaer Depression till the early Devonian. It was uplifted from late Devonian and underwent erosion until the end of the Triassic. From the Jurassic, this area subsided again and accumulated nearly 3000 m of late Mesozoic and Cenozoic sediments to form a separated depression (Wu et al., 2002, 2003).

The Tazhong Low Uplift is a paleo-uplift, which emerged during late Ordovician and received some sediments during the late Silurian to early Devonian. This area rose rapidly during the early Hercynian movement in the late Devonian and suffered from extensive erosion with a loss of 300-500 m (Qiu et al., 1997; Zhang et al., 2000b; Lu et al., 2004). This exposed the Silurian or middle-upper Ordovician succession at the surface in structurally high positions and the Silurian sediments were only preserved in structural lows (Fig. 1B). The Lunnan Low Uplift, located in the middle part of the Tabei Uplift (Fig. 1A), has a similar structural-depositional evolution to the Tazhong Low Uplift. It was eventually formed in the late Devonian when the Silurian-Devonian strata were truncated in structurally high positions, whereas some Silurian was still preserved in the south slope to form stratigraphic traps (Jia, 1999). The variations in structural-sedimentary history between different structural units are probably the major factors controlling the formation and evolution of the Silurian reservoirs.

# 3. Samples and methods

Core samples of the Silurian bituminous sandstones were collected from the Tabei Uplift, Tazhong Low Uplift, eastern Manjiaer Depression and Yingjisu Depression (Fig. 1A). The collected samples were grey to black siltstones or fine-grained sandstones and most of them smell strongly of oil.

Double polished thin sections and one side polished blocks of sandstones were prepared for microscopic examination of fluid inclusions and reservoir bitumen. The petrological examination of solid bitumen and fluid inclusions were performed on a Leica DMRX microscope equipped with a high pressure mercury vapor lamp and Leica H3 filter cube (No. 513827). The H3 filter cube includes an excitation filter of 420-490 nm, a dichromatic filter of 510 nm and a suppression filter of 515 nm. Measurements of fluid inclusion homogenization temperatures were made with a Linkam THMSG 600 heating stage mounted on a Leica DMRX microscope with an attached Linkam TMS 94 control unit. During homogenization temperature measurements, the heating rates of 5 °C/min were used until homogenization was imminent. The temperature was then further raised stepwise by 0.5 °C increments and kept constant for 30 s between each increment.

Solid bitumen reflectance measurements were carried out on the polished sandstone blocks under reflected light using a Leitz microphotometric system, which was calibrated by a Leitz Saphir standard (R = 0.522%). The pho-



Fig. 1. Sketch maps showing location of the studied areas (A) and a cross-section northeast to southwest showing the structural outline (B) (modified from Jia, 1999).

tometer was provided with a pinhole aperture to read a spot with a diameter of 5  $\mu$ m on the sample surface at a wavelength of 546 nm, using a 50×/0.85 objective in oil immersion ( $n_e$  = 1.518).

The saturate fractions from extracts of Silurian sandstones were analyzed on a Finnigan gas chromatograph equipped with fused silica capillary column (ZB 5 column, 30 m  $\times$  0.25 mm i.d.). The GC was kept at the initial temperature of 45 °C for 1 min, then heated to 295 °C at 3 °C/ min and held at this final temperature for 25 min.

# 4. Results and discussions

# 4.1. Characteristics of the bituminous sandstones

# 4.1.1. Geological distribution

The Silurian reservoir is characterized by abundant solid bitumen. Nearly all the discovered bituminous sandstones are located in the paleo-uplifts and their adjacent slopes, such as the Tabei Uplift, the Tazhong Low Uplift and western slope area of the Manjiaer Depression (Fig. 1A). In addition, this type of bituminous sandstone is also sparsely found within the Mandong area and Yingjisu Depression (Fig. 1A).

Lithologically, the Silurian reservoir includes two sets of strata: the upper sandstone seam and the lower sandstone seam, and they are separated by a suite of red mudstones (Fig. 2A). The bituminous sandstone mainly occurs within the lower sandstone seam where it is covered by the red mudstone seam, and occasionally within the upper sandstone seam where the red mudstone is absent or cut through by faults (Fig. 2B). The thickness of bituminous sandstones in different wells varies significantly, ranging from several meters to more than one hundred meters. For example, the thickness of bituminous sandstones is only around 4.0 m in well TZ18, but it reaches to 86 m in well TZ11 in Tazhong Low Uplift and even to 154 m in well HA1 in Tabei Uplift (Fig. 1A). The Silurian reservoir is overlain unconformably by upper Devonian or lower Carboniferous sequences in which no bitumen has been discovered. This indicates that the precursor oil of the bitumen probably was formed and accumulated before the late Devonian. The shapes and textures of the bituminous sandstone in core samples are widely variable. Three types are recognized: blocked, in layers alternating with non-bituminous sandstone, and as veins wedged into non-bituminous sandstones. Generally the average porosity of the bituminous sandstones is larger than the non-bituminous sandstone (Jiang et al., 2006). This implies that the paleo-petroleum fluids preferred to enter into larger inter-granular pores with smaller capillary resistance (Hirsch and Thompson, 1995).

# 4.1.2. Microscopic characteristics of bitumen

The bitumen from the Silurian reservoir sandstone is microscopically identified as pure solid bitumen and bituminous-clay matrix. The former mainly occurs in pores of quartz grains with various shapes, while the latter is amorphous and usually mixed with the clay minerals. Moreover, the pure bitumen includes two types with dis-



Fig. 2. A simplified Silurian stratigraphic column based on the well TZ11 (A), and a cross-section through the Tazhong area showing the distribution of bituminous sandstone (B) (modified from Liu et al., 1996; Yang, 1995, respectively. See well locations in Fig. 1A).

tinct characteristics under reflected and fluorescence lights. Type I exhibits grey colors in reflected light and has no or weak brown fluorescence around bitumen margins with reflectance values of 0.34–0.57% (Fig. 3A and Table 1), whereas Type II has a dark grey color with relatively lower reflectance values of 0.15–0.28% and brown fluorescence (Table 1 and Fig. 3B).

The fluorescence colors of organic matter depend in the first instance on chemical composition, which is controlled not only by maturity but by several other processes (Lin and Davis, 1988; George et al., 2001). For example, biodegradation decreases the aliphatic/aromatic hydrocarbon ratio and increases the proportion of polar components (Bailey et al., 1973), thereby leading to a decrease in the specific gravity of the oil and being expected to alter the fluorescence of residual oils, i.e. reservoir bitumen (George et al., 2001). For example, crude oil in the Hebron I-13 well (Jeanne d'Arc Basin, Canada) is red-shifted compared to inclusion oils, which was interpreted to be due to biodegradation of the crude oil (Stasiuk and Snowdon, 1997). Therefore, the variation in the brown fluorescence colors of a bitumen grain is probably related to the extent of biodegradation in the Silurian reservoirs. It was also observed that the type of bitumen in the bituminous sandstone has a close relationship with the present reservoir petroleum fluids. Where the bituminous sandstone is a commercial oil reservoir, both types of bitumen are developed, such as in wells TZ11 and TZ12, whereas in the bituminous sandstone bearing no movable oils (namely a dry layer) the Type I bitumen with high reflectance is dominant, such as in the well HA1.

# 4.1.3. Oil shows in bituminous sandstones

Three types of movable oils were discovered in the Silurian bituminous sandstone reservoir from the Tazhong area (Table 2). The normal oil and light oil mainly accumulate in the upper sub-seam of the lower sandstone seam, overlain by the red mudstone, while the heavy oil is mainly discovered in the lower sub-seam, covered by a set of thin mudstones (Fig. 2B). Fig. 4 presents three typical gas chromatograms of saturate extracts from bituminous sandstones with different oil stains in Tazhong area, and all of them have a "UCM" hump that indicates an obvious biodegradation. The saturates extracted from a dry bituminous sandstone sample with dominant Type I bitumen are guite rare, which implies that there was a severe biodegradation of earlier oils and no later oil recharged the reservoir (Fig. 4A). However, the situation is different for the sample with both types of bitumen and heavy oil shows (Fig. 4B) and the sample with normal shows (Fig. 4C), and there are saturates superposed on the "UCM" hump with different concentration and distribution mode, indicating obvious later oil charges.

In summary, all the above observations show that the bitumen in the Silurian reservoir genetically belongs to a type of reservoir bitumen associated with biodegradation of paleo-oil pools, and that the Silurian sandstone reservoir experienced multiple petroleum charge events during its



**Fig. 3.** Photomicrographs of pure solid bitumen from the Silurian bituminous reservoir, TZ12, 4374 m (Left, reflected light; Right, fluorescence light). (A) Bitumen with weak fluorescent and a reflectance of 0.34–0.57%; (B) bitumen with fluorescence with a reflectance of 0.15–0.28%.

#### Table 1

Petrological characteristics of solid bitumen in the Silurian reservoir sandstones

| Area         | Well | Description of samples | Depth (m)       | Type I bitumen |                           | Type II bitumen |                           |
|--------------|------|------------------------|-----------------|----------------|---------------------------|-----------------|---------------------------|
|              |      |                        |                 | Fluorescence   | $R_{\rm b}{}^{\rm a}$ (%) | Fluorescence    | $R_{\rm b}{}^{\rm a}$ (%) |
| Tazhong area | TZ4  | Black tar sands        | 3719.00-3738.00 | Dark           | 0.41-0.58/0.51            | Light Brown     | 0.12-0.21/0.18            |
|              | TZ4  | Black tar sands        | 3770.00         | Dark           | 0.34-0.52/0.41            | Brown           | 0.15-0.26/0.21            |
|              | TZ11 | Black tar sands        | 4434.00-4448.00 | Dark           | 0.38-0.47/0.40            | Light Brown     | 0.13-0.24/0.15            |
|              | TZ11 | Black tar sands        | 4448.00-4462.00 | Dark           | 0.41-0.48/0.45            | Brown           | 0.17-0.25/0.22            |
|              | TZ12 | Black tar sands        | 4009.00-4025.00 | Dark           | 0.36-0.57/039             | Light Brown     | 0.14-0.28/0.16            |
|              | TZ12 | Black tar sands        | 4734.14         | Dark           | 0.41-0.67/0.47            | Brown           | 0.21-0.32/0.25            |
|              | TZ10 | Black tar sands        | 4817.50         | Dark           | 0.46-0.64/0.52            | Brown           | 0.15-0.21/0.17            |
|              | TZ30 | Black tar sands        | 4263.00         | Nd             | nd                        | Brown           | 0.19-0.25/0.23            |
|              | TZ31 | Grey tar sands         | 4583.60         | Dark Brown     | 0.32-0.46/0.38            | nd              | nd                        |
|              | TZ31 | Grey tar sands         | 4592.30         | nd             | nd                        | Brown           | 0.18-0.23/0.20            |
|              | TZ33 | Grey tar sands         | 4605.78         | nd             | nd                        | Brown           | 0.24-0.31/0.28            |
| Lunnan area  | HA1  | Black tar sands        | 6080.00         | Dark           | 0.33-0.38/0.35            | Brown           | 0.25-0.29/0.26            |
|              | HA1  | Black tar sands        | 6085.00         | Dark           | 0.34-0.36/0.34            | nd              | nd                        |
|              | HA1  | Black tar sands        | 6088.00         | Dark           | 0.34-0.40/0.36            | nd              | nd                        |
|              | HA1  | Black tar sands        | 6093.00         | Dark           | 0.41-0.48/0.46            | nd              | nd                        |
|              | HA1  | Grey tar sands         | 6161.00         | Dark           | 0.34-0.42/0.38            | nd              | nd                        |
|              | HA1  | Black tar sands        | 6224.00-6242.00 | Dark           | 0.48-0.63/0.57            | nd              | nd                        |
|              | HA1  | Black tar sands        | 6304.00-6320.00 | Dark           | 0.43-0.61/0.55            | nd              | nd                        |
|              | HA1  | Black tar sands        | 6322.00         | Dark           | 0.42-0.57/0.46            | nd              | nd                        |
|              | YM2  | Grey tar sands         | 5024.10         | Dark           | 0.38-0.51/0.43            | Brown           | 0.15-0.26/0.24            |
| Yingjisu Sag | LK1  | Black tar sands        | 4737.55         | Dark           | 0.47-0.49/0.48            | nd              | nd                        |
|              | LK1  | Black tar sands        | 4714.54         | Dark           | 0.42-0.58/0.52            | nd              | nd                        |

<sup>a</sup> Min-max/mean values of bitumen reflectance in oil medium. nd, no data.

#### Table 2

Physical properties of crude oils from the Silurian reservoir sandstone in the Tazhong area

| Oil types  | Density (20 °C) (g/cm <sup>3</sup> ) | Viscosity (mPa s)   | Sulfur content (%) | Polar + asphalt (%) |
|------------|--------------------------------------|---------------------|--------------------|---------------------|
| Heavy oil  | 0.95-1.00                            | 42.00-453.00(80 °C) | 0.66-1.34          | 24.20-39.80         |
| Normal oil | 0.79–0.85                            | 1.03-3.09(50 °C)    | 0.26-0.62          | 1.50-1.60           |
| Light oil  | 0.75-0.76                            | 0.38-0.71(50 °C)    | 0.03-0.06          | 0.54-1.28           |

geological history. In the following sections, we will try to reconstruct the multiple charge history based on fluid inclusions and other available geochemical data.

#### 4.2. Fluid inclusions analysis

#### 4.2.1. Tazhong area

The petroleum inclusions in the bituminous sandstone from the Tazhong area are quite abundant and most of them have two phases (oil and gas). Two groups are identified by their fluorescence colors and homogenization temperatures. The Group I has yellow or brown fluorescence with homogenization temperatures of 60–73 °C and a mean value of 67 °C (Figs. 5A and B and 6A). Their coeval aqueous inclusions homogenize between 65 and 85 °C, with an average of 79 °C. Group II has a green-yellow fluorescence and their homogenization temperatures range from 105 to 130 °C with an average of 117 °C (Figs. 5C and 6A). Their coeval aqueous inclusions have homogenization temperatures of 110–135 °C with an average of 121 °C.

Numerous studies have shown that the fluorescence color of a petroleum inclusion is closely related to its organic components (McLimans, 1987; Stasiuk and Snowdon, 1997; George et al., 2001; Blanchet et al., 2003). Normally, yellow and green colors indicate normal oils, brown or red brown color heavy oils, and blue color light oils. However, this relationship appears to override the use of fluorescence color of petroleum inclusions as a marker of maturity (Pironon and Pradier, 1992; George et al., 2001). This caution is quite significant for basins with multiple source rocks, such as the lapped basins in western China because the compositional variations probably override the maturity constraint on the fluorescence changes. Therefore, the two groups of petroleum inclusions in the Tazhong area only reflect the entrapments of both normal and heavy oils and their maturities need to be confirmed by their molecular geochemistry parameters (George et al., 2001) and their relative charge sequences have to be constrained by other geological and geochemical data.

#### 4.2.2. Lunnan area

Only well JN1 from the Lunnan area penetrated the Silurian bituminous sandstone. There are also two groups of petroleum inclusions in the sandstone (Fig. 5D). The Group I petroleum inclusions exhibit a yellowish fluorescence with homogenization temperatures in the range of 90– 110 °C, and the Group II inclusions have yellow to greenyellow fluorescence and homogenization temperatures of 120–145 °C (Fig. 6B). Their coeval aqueous inclusions have homogenization temperature range of 95–115 °C (average 108 °C) and 125–150 °C (average 132 °C) (Fig. 6B) and both



**Fig. 4.** Gas chromatograms of saturates from three different types of bituminous sandstones. (A) Type I bitumen dominated sample, TZ11, 4422.78–4423.76 m; (B) sample with both type bitumen and heavy oil stains, TZ11, 4432.40 m; (C) sample with normal oil shows, TZ117, 4299.73 m. The gas chromatograms of sample A is amplified in height by two times compared with sample B and C because of extreme low concentration in saturates.

groups of petroleum inclusions show the entrapment of normal oils.

#### 4.2.3. Manxi area

Well YW2 is representative of the Manxi area. Its situation is quite similar to the Lunnan area. There are two groups of petroleum inclusions (Figs. 5E and 6C). Group I inclusions have yellow fluorescence and homogenization temperatures of 58–73 °C, indicating normal oil entrapment, and the Group II inclusions have a variable green-yellow to blue fluorescence with a larger volumetric gas/liquid ratio, and a wide homogenization temperature range of 90–130 °C implying the oils trapped have a wide composition ranging from normal oil to light oil. The mean temperatures of the aqueous inclusions associated with the two groups of petroleum inclusions are 80 and  $120 \,^{\circ}$ C, respectively.

# 4.2.4. Mandong-Yingjisu areas

Well LK 1 was selected in this study to represent this area. Two groups of petroleum inclusions are recognized, one with yellow fluorescence and the other with green or blue-green fluorescence and nearly transparent in transmitted light (Fig. 5F). However, both of them have similar homogenization temperatures in the range 60–85 °C, and coeval aqueous inclusions with homogenization temperatures of 65–90 °C (Fig. 6D). The average homogenization temperatures of the two groups of petroleum inclusions



**Fig. 5.** Photomicrographs of typical petroleum inclusions from the Silurian bituminous reservoirs in different areas. (A) Petroleum inclusions with brown fluorescence, TZ117, 4430.84 m; (B) petroleum inclusions with yellow fluorescence, TZ117, 4430.84 m; (C) petroleum inclusions with yellow-green fluorescence, TZ11, 4418.80 m; (D) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with yellow-green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluorescence, JN1, 5410.20 m; (E) petroleum inclusions with green fluo

and their coeval aqueous inclusions are a little different, 71 and 76 °C for the yellow fluorescence group and 76 and 84 °C for the green fluorescence group. The petrological variance under fluorescence and transmitted lights of the petroleum inclusions indicates that the oils trapped by the two groups of inclusions are quite different, and are interpreted to be normal oil and light oil, respectively. Recent explorations in this area discovered oil cracking gas and the related gas inclusions were trapped during later Tertiary (Xiao et al., 2005). Therefore, the petroleum inclusions associated with cracked oil (or light oil) should be formed before the late Tertiary but after the re-subsidence period of this depression and it is believed that the formation time of petroleum inclusions with yellow fluorescence was earlier than those with green or blue green fluorescence though they have similar homogenization temperatures.

# 4.3. Petroleum charge history in the bituminous sandstones

It is clear from the above discussions that there were petroleum events accompanied by multiple phases of petroleum fluid charge into the Silurian sandstone reservoir. The homogenization temperatures, especially those



**Fig. 6.** Histograms of homogenization temperatures of fluid inclusions in Silurian reservoirs from the four studied areas. (A) Tazhong area; (B) Lunnan area; (C) Manxi area; (D) Mandong–Yingjisu area. Ruled, petroleum inclusions; blank, aqueous fluid inclusions.

of aqueous inclusions coeval with petroleum inclusions are commonly used to predict the time of fluid entrapment by integration with a thermal history, e.g. timetemperature plot (Bodnar, 1990; Aplin et al., 2000; Parnell et al., 2001). The stratigraphic data and paleo-geothermal gradients adopted in this paper are from the Tarim Institute of Petroleum Exploration and Development (2005) and Zhang et al. (2004), respectively. Zhang et al. (2000b) investigated the approximate stratum loss at major unconformities in the studied areas. They found that the eroded thickness of Silurian-lower Devonian sediments was about 1500 m in the Mandong-Yingjisu area and in the range of 300-500 m in the Tazhong and Lunnan areas. Using these data, the formation times of petroleum inclusions in the Silurian sandstone reservoir from the studied areas were deduced from their time-temperature plots (Fig. 7). The timing of the entrapment of inclusions is summarized in Table 3.

The present petroleum in the Silurian sandstone reservoirs mainly originated from the marine organic matters (Liang et al., 2000; Zhang et al., 2000a). The Cambrian source rock has a widespread distribution from the depression to the paleo-uplifts, whereas the mid-upper Ordovician source rock developed only in the slope areas of paleo-uplifts, especially in the Tazhong Low Uplift and Lunnan Low Uplift (Liang et al., 2000; Lu et al., 2004). Fig. 8 presents the thermal evolution of source rocks in the slope areas of the above two uplifts where both sets of source rocks have been well documented (Liang et al., 2000; Zhang et al., 2004). The oil window of the Cambrian source rocks covered a wide time span from late Ordovician to Triassic because of the frequent uplift and erosion



**Fig. 7.** Plots showing the average formation times of petroleum inclusions in the Silurian reservoirs. (A) Well TZ11 in the Tazhong area; (B) well YW2 in the Manxi area; (C) well JN1 in the Lunnan area; (D) well LK1 in the Mandong–Yingjisu area. The time–temperature curves are calculated according to the sample average depths and average homogenization temperatures of aqueous fluid inclusions associated with petroleum inclusions.

events, and the present maturity extends to the wet gas to dry gas stage (Zhang et al., 2004). However, the mid-



**Fig. 8.** Plots showing the evolution of thermal maturity for the mid-lower Cambrian and the mid-upper Ordovician source rocks using the Easy Ro model of Sweeney and Burnham (1990) based on the geothermal history of Zhang et al. (2004). (A) Well TZ12 in the slope areas of Tazhong Low Uplift; (B) well LN45 in the slope area of Lunnan Low Uplift.

upper Ordovician source rocks have a low thermal maturity and they were marginally mature during the period from Permian to Cretaceous and only began to mature during the Tertiary and are still within the oil window. Liang et al. (2000) stated that it was just because of this low maturity of source rocks in the slopes that there are abundant commercial oils in these areas. Zhang et al. (2004) also confirmed that these commercial oils are mainly from this set of low maturity of mid-upper Ordovician source rocks. Therefore, there were three major episodes of petroleum generation and expulsion around the paleo-uplifts that are nearly continuous in time and space. The first is related to mid-lower Cambrian source rocks in the deeper parts around paleo-uplifts during the Ordovician to early Devonian, the second is mainly associated with shallower parts of the mid-lower Cambrian source rocks around paleo-uplifts during the Carboniferous to Triassic, and the third is mainly related to the mid-upper Ordovician source rocks around the uplifts that controlled the present commercial oil pools during after the Tertiary.

By considering the geological background of the studied areas and the formation times and the petrological

Table 3

A summary and comparison of trapping time of petroleum inclusions from the Silurian reservoir in different areas

| Area (Well)  | Tazhong (Well TZ11) | Manxi (Well YW2) | Lunnan (Well JN1) | Mandong (Well LK1) |
|--|---------------------|------------------|-------------------|--------------------|
| Time for group I petroleum inclusions entrapment (Ma)  | -225                | -122             | -20               | -386               |
| Time for group II petroleum inclusions entrapment (Ma) | -5                  | -12              | -10               | -130               |

characteristics of the petroleum inclusions in the bituminous sandstones, three petroleum fluid charge events can be distinguished:

Event I: Early Devonian with normal oils trapped at temperatures of about 60–80 °C;

Event II: Mainly during Triassic–Cretaceous with different types of oil trapped at average temperatures between 80–85 °C; and

Event III: During the late Tertiary with normal and light oils trapped at average temperatures of  $110-135 \circ C$ .

It should be pointed out that although the two groups of petroleum inclusions from the Lunnan Low Uplift (well JN1) were included in Event III, it is possible they could be related to different sources. According to Liu et al. (2003), the Caohu Sag, located in the east of the Lunnan Low Uplift (Fig. 1A), was probably another petroleum source in addition to the Manjiaer Depression.

However, petroleum inclusions originating from the three identified events have not been observed together in any of the four studied areas owing to the constraints imposed by their source rocks and trapping conditions. Event I indicates the charge time of the precursor oil of bitumen. They were found in the Silurian sandstone reservoir only from the Mandong-Yingjisu area where the Silurian sandstone was buried deeply enough for petroleum inclusions to be formed at that time. Liu et al. (2003) reported petroleum inclusions from this event in the Ordovician carbonate reservoirs from the Lunnan area. Event II petroleum inclusions were widespread and trapped over a long period of geological time. They were recorded in all the other three studied areas except the Lunnan area. Event III petroleum inclusions have not been found in this study for the Mandong–Yingjisu area, but Xiao et al. (2005) reported gas inclusions that are related to the upward migration of oil cracking gas from Cambrian and Ordovician strata during Tertiary. As petroleum inclusions in reservoirs record the history of petroleum migration and accumulation (Mclimans, 1987; Parnell et al., 1996, 2001), the petroleum inclusions in the Silurian reservoir can be used to infer that there were three petroleum charge events, which occurred during early Devonian, Triassic-Cretaceous and late Tertiary.

# 4.4. Formation and evolution of oil pools

Based on the above discussion, the formation and evolution of the Silurian oil pools can be divided into three stages:

Stage I: The formation of the paleo-oil pools. During the early Devonian, the Cambrian source rocks from the deeper parts of paleo-uplifts slopes reached their main stage of oil generation and the generated oil migrated toward the paleo-uplifts and slope areas along faults and unconformities into the Silurian sandstone to form oil pools covering a very extensive area. Most of the oil pools, however, were eroded or biodegraded during the late Devonian associated with the early Hercynian Orogeny and eventually evolved to solid bitumen due to severe biodegradation thereby forming residual paleo-oil pools. This charge event was well recorded by petroleum inclusions formed at the time of 386 Ma in well LK1 in Mandong area. Gong et al. (2007) also identified this charge event in the Tabei Uplift based on the molecular composition analysis of inclusion oil in the Ordovician reservoirs.

Stage II: The alteration of the paleo-oil pools. The Cambrian source rock from the shallower part of paleouplifts reached oil generation peak around Permian to Triassic and the mid-upper Ordovician source rocks were marginally mature during this time. These oils with different maturity and origins recharged some of the Silurian bituminous sandstone reservoirs. As the bitumen in the reservoir was in the low-mature stage during this period, it was altered by the recharged oil to form the low reflectance bitumen and a type of heavy oil. This process is documented by the occurrences of petroleum inclusions and low reflectance bitumen with dominant brown fluorescence although some normal oils with vellow fluorescence were trapped as inclusions. Because of the complexity of oil sources and interaction between recharged oil and early bitumen, the observed petroleum inclusions are quite variable and complicated in fluorescence and formation time.

Stage III. The accumulation of commercial oil pools. During the late Tertiary, the whole Tarim basin resubsided rapidly and the mid-upper Ordovician source rock in the slope areas of paleo-uplifts entered into the oil window and the generated oil accumulated in the Silurian reservoirs to form the present commercial oil pools. As the mid-upper Ordovician source rock experienced a wide range of maturation, from mature to highly mature, in different locations, it was possible for the oil pools to be charged with oils in the normal and light oil range. The petroleum inclusions formed during this period have mainly fluorescence with yellow to green-yellow, as the oil entrapped in inclusions has not suffered from biodegradation that often leads to a red shift of fluorescence (George et al., 2001).

It should be noted that this is a general model for the evolution of the paleo-oil pools and variations appropriate to different areas may be required due to differences in their structural evolution and source rock maturation levels. The three stages were quite typical of the Tazhong Low Uplift where both mid-lower Cambrian and mid-upper Ordovician source rocks were well developed (Liang et al., 2000).

# 5. Conclusions

Combining the petrological and geochemical characteristics of the Silurian bituminous sandstone with the geological background of the study area where the Manjiaer petroleum system was developed, it has been possible to conclude the following:

- (1) There are four types of oil shows in the reservoir: severely biodegraded solid bitumen, biodegraded heavy oil, normal oils and light oils with little biodegradation. There is evidence of three groups of petroleum inclusions in the Silurian bituminous sandstones, indicating that there were three petroleum charge events around the depression and paleo-uplifts which occurred during the early Devonian, Triassic-Cretaceous and late Tertiary.
- (2) Three episodes of oil formation and migration were identified by combining the petroleum inclusions and thermal evolution of possible source rocks. The petroleum charge event I was related to the midlower Cambrian source rock in the deeper parts of paleo-uplifts and shallower parts of the Manjiaer Depression, event II to both the shallower mid-lower Cambrian and the deeper mid-upper Ordovician source rocks in paleo-uplifts and slope areas, and event III mainly to the mid-upper Ordovician source rock in the slope areas of paleo-uplifts.
- (3) The oil pools formed during event I were eroded or altered by biodegradation in late Devonian due to the uplift associated with early Hercynian Orogeny and eventually formed huge residual paleo-oil pools. During event II petroleum fluids recharged the paleo-oil pools and interacted with the earlier bitumen to form a type of heavy oil and soft bitumen with low reflectance. The event III made the dominant contribution to the current commercial oil pools in the reservoir.

#### Acknowledgements

The authors are indebted to Professor D.G. Liang and two anonymous reviewers for their insightful comments and suggestions that significantly improved the manuscript. The work was financially supported by the Natural Science Fund for Distinguished Young Scholars (Grant No. 40625011) and the Key Project of Chinese Academy of Science (Grant No. KZCX2-YW-114-2).

#### Associate Editor-Digang Liang

# References

- Aplin, A.C., Larter, S.R., Bigge, M.A., Macleod, G., Swarbrick, R.E., Grunberger, D., 2000. PVTX history of the North Sea's Judy oilfield. Journal of Geochemical Exploration 69–70, 641–644.
- Bailey, N.J.L., Krause, H.R., Evans, C.R., Rogers, M.A., 1973. Alteration of crude oil by waters and bacteria – evidence from geochemical and isotope studies. American Association of Petroleum Geologists Bulletin 57, 1276–1290.
- Blanchet, A., Pagel, M., Walgenwitz, F., Lopez, A., 2003. Microspectrofluorimetric and microthermometric evidence for variability in hydrocarbon inclusions in quartz overgrowths: implications for inclusion trapping in the Alwyn North field, North Sea. Organic Geochemistry 34, 1477–1490.
- Bodnar, R.J., 1990. Petroleum migration in the Miocene Monterey Formation, California, USA: constraints from fluid inclusion studies. Mineralogical Magazine 54, 295–304.
- George, S.C., Ruble, T.E., Dutkiewicz, A., Eadington, P.J., 2001. Assessing the maturity of oil trapped in fluid inclusions using molecular geochemistry data and visually-determined fluorescence colors. Applied Geochemistry 16, 451–473.

- Gong, S., George, S.C., Volk, H., Liu, K., Peng, P.A., 2007. Petroleum charge history in the Lunnan Low Uplift, Tarim Basin, China – evidence from oil-bearing fluid inclusions. Organic Geochemistry 38, 1341–1355.
- Guo, R.T., Xiao, X.M., Wang, J.B., 2001. Characteristics and description of hybrid petroleum system – a case study of the Tazhong petroleum system of the Tarim Basin. Geochimica 30, 425–432. (in Chinese).
- Hirsch, L.M., Thompson, A.H., 1995. Minimum saturations and buoyancy in secondary migration. American Association of Petroleum Geologists Bulletin 79, 696–710.
- Jia, C.Z., 1999. The structural characteristics and hydrocarbon accumulation in Tarim Basin. Xinjiang Petroleum Geology 20, 177– 183. (in Chinese).
- Jiang, Z.X., Pang, X.Q., Wang, X.D., Zhang, J., Li, H.Y., 2006. Determination methods of effective thickness of Silurian bituminous sandstones in the Tarim Basin. Acta Geologica Sinica 80, 418–423. (in Chinese).
- Kang, Y., Kang, Z., 1996. Tectonic evolution and oil and gas of Tarim basin. Journal of Southeast Asian Earth Sciences 13, 317–325.
- Li, D.S., Liang, D.G., Jia, C.Z., Wang, G., Wu, Q.Z., He, D.F., 1996. Hydrocarbon accumulations in the Tarim Basin, China. American Association of Petroleum Geologists Bulletin 80, 1587–1603.
- Liang, D.G., Zhang, S.C., Zhang, B.M., Wang, F., 2000. Understanding on marine oil generation in China based on Tarim Basin. Earth Science Frontier 7, 534–547. (in Chinese).
- Lin, R., Davis, A., 1988. A fluorogeochemical model for coal macerals. Organic Geochemistry 12, 363–374.
- Liu, D.H., Xiao, X.M., Mi, J.K., Li, X.Q., Shen, J.K., Song, Z.G., Peng, P.A., 2003. Determination of trapping pressure and temperature of petroleum inclusions using PVT software – a case study of Lower Ordovician carbonates from the Lunnan Low Uplift, Tarim Basin. Marine and Petroleum Geology 20, 29–43.
- Liu, L.F., Zhao, J.Z., Zhang, S.C., Fang, J.H., Xiao, Z.Y., 2001. The depositional and structural settings and the bituminous sandstone distribution characters of the Silurian in Tarim basin. Acta Petrolei Sinica 21, 11– 17. (in Chinese).
- Liu, S.P., Zhong, G.F., Liu, X.F., Pen, D.T., 1996. Characteristics and evaluation of Silurian sandstone reservoir in Tazhong area, Tarim. Journal of the Jianghan Petroleum Institute 18, 21–25. (in Chinese).
- Lomando, A.J., 1992. The influence of solid reservoir bitumen on reservoir quality. American Association of Petroleum Geologists Bulletin 76, 1137–1152.
- Lu, X.X., Jin, Z.J., Liu, L.F., Xu, S.L., Zhou, X.Y., Pi, X.J., Yang, H.J., 2004. Oil and gas accumulations in the Ordovician carbonates in the Tazhong Uplift of Tarim Basin, west China. Journal of Petroleum Science and Engineering 41, 109–121.
- McLimans, R.K., 1987. The application of fluid inclusions to migration of oil and diagenesis in petroleum reservoirs. Applied Geochemistry 2, 585–603.
- Parnell, J., Carey, P.F., Monson, B., 1996. Fluid inclusion constraints on temperatures of petroleum migration from authigenic quartz in bitumen veins. Chemical Geology 129, 217–226.
- Parnell, J., Middleton, D., Chen, H.H., Hall, D., 2001. The use of integrated fluid inclusions studies in constraining oil charge history and reservoir compartmentation: examples from the Jeanne d'Arc Basin, offshore Newfoundland. Marine and Petroleum Geology 18, 535–549.
- Pironon, J., Pradier, B., 1992. Ultraviolet-fluorescence alteration of hydrocarbon fluid inclusions. Organic Geochemistry 18, 501–509.
- Qiu, N., Jin, Z., Wang, F., 1997. The effect of the complex geothermal field based on the multi-structure evolution to hydrocarbon generation – a case of Tazhong area in Tarim Basin. Acta Sedimentologica Sinica 15, 142–144. (in Chinese).
- Stasiuk, L.D., Snowdon, L.R., 1997. Fluorescence micro-spectrometry of synthetic and natural hydrocarbon fluid inclusions: crude oil chemistry, density, application to petroleum migration. Applied Geochemistry 12, 229–241.
- Sweeney, J.J., Burnham, A.K., 1990. Evaluation of a simple model of vitrinite reflectance based on chemical kinetics. American Association of Petroleum Geologists Bulletin 74, 1559–1570.
- Tarim Institute of Petroleum Exploration and Development, 2005. The latest stratigraphic data of exploration and evaluation wells in Tarim Basin. Tarim Oilfield Company internal report (in Chinese).
- Wu, G.G., Li, H.Q., Chu, B.J., Xia, B., Wang, H., Pu, G.M., 2002. Geotectonic evolution and petroleum accumulation in east Tarim. Geotectonic et Metallogenia 26, 229–234. (in Chinese).
- Wu, G.G., Xia, B., Wang, H., Cheng, Z.Y., 2003. The characteristics and partitioning of tectonic deformation in east Tarim Basin. Xinjiang Geology 18, 161–165. (in Chinese).

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- Xiao, Z., Lu, Y., Wu, Y., Zhao, F.Y., Qian, L., Jin, Y.A., 2005. A preliminary study on natural gas genesis and pool-forming period in the Silurian reservoir of well Mandong 1, east Tarim Basin. Chinese Journal of Geology 34, 155–160. (in Chinese).
- Yang, H.J., 1995. The evaluation of Silurian reservoirs of the northern slope in Tazhong area, Tarim Basin. Tarim Oilfield Company internal report (in Chinese).
- Zhang, M., Zhang, J., Mao, Z.C., Zhao, H.J., Hu, B.L., 2002. The hydrocarbon compositional heterogeneity of petroleum reservoir and its geological significance in Tazhong area. Fault-Block Oil and Gas Field 9, 5–8. (in Chinese).
- Zhang, S.C., Hanson, A.D., Moldowan, J.M., Graham, S.A., Liang, D.G., Chang, E., Fago, F., 2000a. Palaeozoic oil-source rock correlations in the Tarim basin, NW China. Organic Geochemistry 31, 273– 286.
- Zhang, S.C., Liang, D.G., Zhang, B.M., 2004. The Formation of Marine Oil and Gas in the Tarim Basin, first ed. Petroleum Industry Press, Beijing. (in Chinese).
- Chang, Y.W., Jin, Z.J., Liu, G.C., Li, J.C., 2000b. Study on the formation of unconformities and the amount of eroded sedimentation in Tarim basin. Earth Science Frontiers 7, 449–457. (in Chinese).