Determination of Time of Oil Cracking into Gas in Weiyuan Paleo-Oil Pool in Sichuan Basin, South China

Yusong Yuan*, Dongsheng Sun, and Juanhua Lin
Petroleum Exploration and Production Research Institute, SINOPEC, Beijing, China

ABSTRACT
Weiyuan gas field, located in the Sichuan basin of south China, is a large marine gas field with the oldest reservoir (the Sinian sequences) in south China. The hydrocarbon origin of the gas field has long been debated by petroleum geologists. Recently, it was recognized that a paleo-oil pool maybe the significant contributor to the gas field. Consequently, when the paleo-oil pool had been cracked into gas has become an interesting topic. Actually, the time determination of oil cracking into gas is of significance to marine gas reservoir exploration in south China, because it is a pervasive hydrocarbon resource of marine gas reservoirs in south China. Characteristics of the gas composition, fluid inclusion, and bitumen in Sinian reservoir show that the present-day Weiyuan gas field originates from a paleo-oil pool cracked at high temperature. Data collected from the previous documents show that the temperature window of oil cracking into gas is around 160 to 210 °C on a geological time scale. On the basis of vitrinite reflectance, the heat flow history and erosion thickness in Weiyuan area were reconstructed. With the combination of heat flow history and burial history, the temperature history the paleo-oil pool experienced was reconstructed. The time of the paleo-oil cracking into gas is determined according to the oil cracking temperature window. The results show that the oil in the Weiyuan Sinian paleo-oil pool cracked into gas during the late Triassic to Eocene age.

Keywords: Sichuan Basin, Weiyuan Paleo-oil Pool, Oil Cracking into Gas, Oil Cracking Temperature Window, Heat Flow History

INTRODUCTION
Marine petroleum in sedimentary basins of south China has many types of origin such as kerogen, bitumen, organic acid salt, scattered hydrocarbon, crude oil in paleo-oil pools, etc. [1, 2]. As the marine sequences in south China experienced multi-stages of uplift and subsidence, they generally underwent multi-stages of hydrocarbon generation such as early and secondary hydrocarbon generation. Thus hydrocarbon in south China is characterized by multi-origins and multi-stages [3, 4]. Among the multi-origins, the crude oil cracking is one of the most important origins for the gas reservoirs in south China’s marine carbonate sequences [5]. The marine carbonate sequences are normally located in a lower position of the superimposition sedimentary basins in south China and, therefore, have undergone a relatively higher paleo-temperature. At a high temperature, crude oil in paleo-oil pools could be cracked into gas. Many of the discovered large gas reservoirs in south China originated from crude oil cracking including Weiyuan, Puguan, and Yuanba gas fields in Sichuan basin.
Basin [1, 6]. To correctly determine the history of gas reservoir formation, including the origin of gas, it is necessary to make the oil and gas accumulation controls clear in south China marine carbonate sequences [4]. As gas cracked from crude oil is an important source of marine gas in south China, how to determine the time of oil cracking in a paleo-oil pool is of significance to the exploration in the marine sequences. There are well-established methods of history reconstruction of hydrocarbon generation for kerogen [7], but there are no methods for oil cracking. In this paper, we proposed a method of determining the time of oil cracking called “oil cracking temperature window method” and took Weiyuan Sinian paleo-oil pool as a case study.

Geological Settings

The sedimentary stratigraphy of Sichuan basin consists of Sinian to Quaternary rocks (Figure 1), including the Sinian-middle Triassic marine deposits in cratonic basin and the late Triassic-Cenozoic fluvial-lacustrine sediments in foreland basins [8, 9]. The Dengying formation of the upper Sinian includes mainly dolomite and shale interbeds. In contrast, the Jiulaodong formation of the lower Cambrian is composed of interbed black and gray green shales deposited in open marine-continental shelf settings. They acted as reservoir and source rocks respectively in the lower Paleozoic petroleum system [9].

The Sichuan basin has experienced several episodes of tectonic movements [10, 11], including Caledonian, Hercynian, Indosinian, Yanshanian, and Himalaya tectonic movements. Among them, the Caledonian movements resulted in the formation of the Leshan-Longnüsi paleo-uplift, which is also called central Sichuan paleo-uplift [12]. This paleo-uplift was initially formed during the late Sinian-Cambrian, was developed in the Ordovician, and was fully formed in the Silurian. It has been a giant uplift controlled by basement faults. As calculated by the area, where the Silurian sequences are missed, it covers an area of 6.25×10⁴ km² [13].

Weiyuan Paleo-oil Pool

In Weiyuan county and Rong county, Sichuan province, China, the oldest gas filed in China in the marine carbonate sequences was found in 1964, which called Weiyuan Sinian gas field [14]; it is the only one up to now in China, whose reservoir is the Sinian carbonate. The regional paleostructure revolution, natural gas component, and reservoir bitumen show that the gas is originated from oil cracking [15, 16]. That is to say that the present-day Weiyuan gas field was an oil pool in the past.

The regional paleostructure revolution provided geological advantages for the formation of the Weiyuan paleo-oil pool [10]. The tectonic evolution of Leshan-Longnüsi paleo-uplift located in the central-western Sichuan basin, as a large positive structural unit of the basin, has an important influence on the formation and distribution of the Sinian gas reservoirs. The Weiyuan area was located in the slope of the central Sichuan uplift in the early stage and located in the core of the uplift in the late period. Therefore, it has been a favorable area for oil and gas migration and accumulation. The source rocks had been matured to hydrocarbon generation stage in the southern and southeastern Sichuan basin, when the central Sichuan uplift was formed. The uplifting movement at the end of the Sinian provided favorable geological conditions such as weathering and solution to form high quality reservoirs. Moreover, traps were also formed, when the paleo-uplift was forming and the top seal, which made of the Jiulaodong formation dominated by mudrocks, had a sealing capacity for oil [17]. Sufficient hydrocarbon sources, high quality reservoirs, good top seal, advantageous structural position, and trap conditions facilitated the formation of Weiyuan paleo-oil pool.
The paleo-oil pool covered as large an area as 1900 km$^2$ in the Paleozoic time [16]. The gas component in the present-day Weiyuan gas field is characterized by oil cracking gas [16]. Pyrolysis experiments in close system find out the differences between the kerogen pyrolysis gas and oil cracking gas. With increasing maturity, the ratio of C$_1$ to C$_2$ of the kerogen pyrolysis in gas increases, but the ratio of C$_2$ to C$_3$ remains constant; however, the ratio of C$_1$ to C$_2$ of the oil
cracking gas remains constant, but the ratio of the $C_2$ to $C_3$ increases. Therefore, the natural gas from kerogen pyrolysis or from oil thermal cracking can be distinguished by the gas component board chart of $\ln(C_1/C_2)$ and $\ln(C_2/C_3)$ [18]. The Sinian gas component data from Weiyuan gas field fall into the oil cracking gas area in the $\ln(C_1/C_2)$ and $\ln(C_2/C_3)$ board chart [16]. Therefore, the gas in the Weiyuan Sinian reservoir is originated from crude oil cracking.

Moreover, the bitumen in the Weiyuan Sinian reservoir is the oil cracking product. The bitumen in the Sinian reservoir is found in almost all the boreholes located in the central Sichuan uplift drilled to the Sinian reservoir; also, in the higher position of the uplift, higher content of the bitumen is found [16]. The bitumen reflectance (Rb) in the Weiyuan Sinian reservoir is generally higher than 2.5%. For example, the Rb of the 4th, 3rd, and second members of the Dengying formation are 2.76%, 2.71%, and 2.72% respectively [19]. The high Rb in the reservoir is a direct evidence of oil cracking into gas [15, 20].

### Oil Cracking Temperature Window

Crude oil from kerogen, whether resided in source rock or migrated into a trap, will crack into gas at certain elevated temperatures. Previous work provided general cognitions about the issue of oil cracking into gas. Oil cracking into gas is dominated by temperature and influenced by oil type, oil component, reservoir pressure, formation water, and mineral [21, 25].

A collection of oil cracking into gas temperature data from the previous work are summarized in Table 1. It can be seen from Table 1 that the oil cracking into gas temperature is roughly between 160 to 210 °C. Therefore, the temperature range of 160 to 210 °C was chosen as the oil cracking temperature window in this paper.

<table>
<thead>
<tr>
<th>Oil field/Basin</th>
<th>Preliminary cracking temperature (°C)</th>
<th>Fully cracking temperature (°C)</th>
<th>Data origin</th>
</tr>
</thead>
<tbody>
<tr>
<td>Great valley, California</td>
<td>130-150</td>
<td></td>
<td>[26]</td>
</tr>
<tr>
<td>An oil field in Italy</td>
<td>153</td>
<td></td>
<td>[27]</td>
</tr>
<tr>
<td>Willis Benton Basin, United States</td>
<td>182</td>
<td></td>
<td>[21]</td>
</tr>
<tr>
<td>southern Sahara basin, Algeria</td>
<td>130</td>
<td></td>
<td>[28]</td>
</tr>
<tr>
<td>Southwestern Alabama</td>
<td>130-140</td>
<td></td>
<td>[29]</td>
</tr>
<tr>
<td>Horse HaKan Delta</td>
<td>140</td>
<td></td>
<td>[30]</td>
</tr>
<tr>
<td>Niger Delta</td>
<td>150</td>
<td></td>
<td>[30]</td>
</tr>
<tr>
<td>Tarim Basin</td>
<td>160</td>
<td>&gt;150</td>
<td>[31]</td>
</tr>
<tr>
<td></td>
<td>160</td>
<td></td>
<td>[32]</td>
</tr>
<tr>
<td>Dongying depression</td>
<td>160</td>
<td>180-225</td>
<td>[34]</td>
</tr>
<tr>
<td></td>
<td>150-180</td>
<td></td>
<td>[35]</td>
</tr>
<tr>
<td>UK Central, North Sea</td>
<td>&gt;174</td>
<td>150</td>
<td>[36]</td>
</tr>
<tr>
<td>Norwegian Continental Shelf</td>
<td>160-190</td>
<td>200</td>
<td>[37]</td>
</tr>
<tr>
<td>Dongying sag, Bohai bay basin</td>
<td>150</td>
<td>200</td>
<td>[24]</td>
</tr>
<tr>
<td>North Sea</td>
<td>170</td>
<td>215</td>
<td>[23]</td>
</tr>
<tr>
<td></td>
<td>150</td>
<td>190</td>
<td>[37]</td>
</tr>
<tr>
<td></td>
<td>150</td>
<td>220</td>
<td>[38]</td>
</tr>
<tr>
<td></td>
<td>150</td>
<td>200</td>
<td>[39]</td>
</tr>
</tbody>
</table>
Oil Cracking History of Weiyuan Paleo-oil Pool

Method for the Reconstruction of Oil Cracking History

Methods about the history reconstruction of hydrocarbon generation for kerogen pyrolysis have fully been developed. Vitrinite reflectance (Ro) calculation according to chemical kinetic models such as the EASY% Ro model can be used [7]; however, no methods have been reported for oil cracking up to now. We proposed a method called “oil cracking temperature window method” to reconstruct the history of oil cracking in a reservoir. Taking the oil reservoir as the study object to substitute for the source rock object, hydrocarbon generation history from kerogen pyrolysis was reconstructed. Firstly, reservoir temperature history was reconstructed based on its burial and regional heat flow histories; then, the time of oil cracking was determined by the combination of reservoir temperature history with oil cracking temperature window. The technology diagram of oil cracking temperature window method is shown in Figure 2.

Figure 2: Technology flow chart showing the oil cracking temperature window method.

Reconstruction of Erosion Thickness and Heat Flow History in Weiyuan Area

To reconstruct burial and temperature histories, the erosion thickness and heat flow history must be reconstructed at first. There are more than 100 boreholes drilled in Weiyuan area. Among them, there are three boreholes which have met pre-Sinian granite rocks (W15, W28, and W117 respectively). The W28 not only has met all the sedimentary sequences in Weiyuan area, but also is an industrial gas well [40]. Therefore, we selected the W28 as a representative well for the reconstructions of erosion thickness, burial, and heat flow histories in order to provide geological boundaries for the determination of oil cracking into gas history.

Methods for the reconstruction of erosion thickness fall into four categories [41]: geothermics methods based on paleotemperature index of vitrinite reflectance (Ro), apatite fission track (AFT), zircon fission track (ZFT), or fluid inclusion (FI); geology methods are based on stratigraphical comparison, strata thickness trend, sedimentation ratio, or sedimentary wave analysis; geophysics methods are based on porosity or acoustic travel time; geochemistry methods are based on the equilibrium concentration of natural gas or Cosmogenic nuclides. Among them, the geothermics method is the frequently used one for the quantitative reconstruction of erosion thickness. We obtained 24 Ro values from Paleozoic and Mesozoic sequences in W28, which is a set of systematic Ro values (Table 2). Therefore, the geothermics method is used in this study.

Vitrinite originating from higher plants is absent in the lower Paleozoic sequences. Ro data of the lower Paleozoic strata in Table 2 was calculated from the reflectance of vitrinite-like maceral (VLMRo) using the below equation established by Xiao et al. (2003).

\[ VRO(\%) = 0.81 \times VLMRo + 0.18 \]  

(1)
Table 2: Vitrinite reflectance data from different strata of Wei 28 well.

<table>
<thead>
<tr>
<th>Depth (m)</th>
<th>VRo (%)</th>
<th>Strata</th>
<th>Depth</th>
<th>VLMRo (%)</th>
<th>VRo (%)</th>
<th>Strata</th>
</tr>
</thead>
<tbody>
<tr>
<td>210</td>
<td>0.78</td>
<td>Middle</td>
<td>1880</td>
<td>2.17</td>
<td>1.94</td>
<td></td>
</tr>
<tr>
<td>310</td>
<td>1.06</td>
<td>Triassic</td>
<td>1990</td>
<td>2.35</td>
<td>2.08</td>
<td></td>
</tr>
<tr>
<td>645.4</td>
<td>1.59</td>
<td></td>
<td>2080</td>
<td>2.39</td>
<td>2.11</td>
<td>Lower Ordovician</td>
</tr>
<tr>
<td>706.73</td>
<td>1.53</td>
<td></td>
<td>2480</td>
<td>3.2</td>
<td>2.77</td>
<td></td>
</tr>
<tr>
<td>710</td>
<td>1.32</td>
<td></td>
<td>2580</td>
<td>2.84</td>
<td>2.48</td>
<td>Lower Cambrian</td>
</tr>
<tr>
<td>910</td>
<td>1.64</td>
<td></td>
<td>2781</td>
<td>3.32</td>
<td>2.87</td>
<td></td>
</tr>
<tr>
<td>1067.9</td>
<td>1.68</td>
<td>Lower</td>
<td>2880</td>
<td>3.28</td>
<td>2.84</td>
<td></td>
</tr>
<tr>
<td>1210</td>
<td>1.58</td>
<td>Triassic</td>
<td>2980</td>
<td>4.01</td>
<td>3.43</td>
<td></td>
</tr>
<tr>
<td>1310</td>
<td>1.76</td>
<td>Upper</td>
<td>3080</td>
<td>3.96</td>
<td>3.39</td>
<td>Permian</td>
</tr>
<tr>
<td>1410</td>
<td>1.84</td>
<td>Permian</td>
<td>3394.25</td>
<td>3.69</td>
<td>3.17</td>
<td></td>
</tr>
<tr>
<td>1590</td>
<td>1.58</td>
<td>Lower</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>Permian</td>
</tr>
<tr>
<td>1690</td>
<td>1.94</td>
<td></td>
<td>-</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>1738.11</td>
<td>1.70</td>
<td>Permian</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>1780</td>
<td>1.85</td>
<td></td>
<td>-</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
</tbody>
</table>

Based on the geothermics method and by means of “Thermodel for windows” software [42], the calculated erosion thickness in W28 is 2889 m constrained by the obtained 24 Ro values (Figure 3). The estimation of the erosion thickness (2889 m) can be reasonable. From the preserved strata, it can be speculated that a large amount of deposits eroded away in Himalayan period in Weiyuan area. The outcrop is Xujiahe formation, upper Triassic in W28. The removed sequences include Paleogene, Cretaceous, and Jurassic. The Xujiahe formation was nearly eroded away, which is just preserved 76 m. In addition, among the 24 Ro values from the W28, the one from the top sample buried 210 m is 0.78%, which is much higher than that from near surface samples. This indicates that the sample buried 210 m in the present-day was buried much deeply in the past. Its over lain sequences on a several-kilometer scale have been eroded away. In Figure 3, G_{present}, is the present-day geothermal gradient; the black solid line shows the present-day temperature profile and G_{paleo} is paleo-geothermal gradient; the red dash line shows the paleo-temperature profile and the blue dots depict the paleo-temperature calculated from Ro data; Ze is the thickness of the removed formations. Cam, O1, P1, P2, T1, T2 and T3 denote Cambrian, lower Ordovician, lower Permian, upper Permian, lower Triassic, middle Triassic, and upper Triassic respectively.

Figure 3: Denudation in well 28, Weiyuan, Sichuan basin.

Paleo-heat flow reconstruction based on the 24 Ro values given above, which are also obtained by means of “Thermodel for windows” software, shows that Weiyuan area reached its peak heat flow in the end of the late Permian (258 Ma bp) and the peak surface paleo-heat flow was as high as 85.3 mW/m^2. It decreased to 70 mW/m^2 at the end of the middle Triassic, and finally dropped to 67 mW/m^2 at the end of the
Paleogene. The present-day surface heat flow is about 67 mW/m² (Figure 4, left graph). From the Figure 4 (b), it can be inferred that the calculated Ro values are in good agreement with the measured ones. This indicates that the heat flow history path (Figure 4) can be accepted.

Figure 4: Paleo-heat flow history of well 28, Weiyuan, Sichuan basin. The modeled heat flow (a) was obtained by inversion using vitrinite reflectance data (b).

Sinian Reservoir Temperature and Oil Cracking Histories

Based on the reconstruction of erosion thickness, the burial history can be reconstructed making use of back-stripping method [43]. Then, the strata temperature history can be reconstructed by the combination of the burial history with strata temperature history. Taking the Sinian reservoir in W28 as a case study, the simulated results show that the Sinian deposits experienced burial depth and temperature increasing in the early Paleozoic. The bottom of the Sinian reservoir reached the burial depth of 3155 m at a temperature of 115 °C at the end of the early Paleozoic. The Devonian-Carboniferous period is an uplifting cooling period without deposits. From the Permian, subsiding and therefore the Sinian reservoir temperature increasing occurred again. In the process of subsiding and temperature increasing, the Sinian reservoir bottom depth reached 4358 m and its temperature rose to 160 °C at the beginning of the late Triassic. The Sinian reservoir top depth reached 6132 m and its temperature increased to 210 °C at the end of the Eocene. The homogenization temperature of fluid inclusion from the upper Sinian carbonate reservoir in Weiyuan and Ziyang area is up to 200 °C [44]. It agrees with the temperature calculated from heat flow and burial histories. Thus, based on the burial history, the temperature history of the reservoir and oil cracking temperature window, it is clear that the time of the crude oil cracking into gas in the Weiyuan Sinian paleo-oil pool is from the late Triassic to the Eocene (upper limit time) (see Figure 5).

Figure 5: History of the oil cracking into gas in the Sinian reservoir of well 28 in Weiyuan area, Sichuan basin; purple line for the Sinian reservoir bottom; black line for the Sinian reservoir top.

Here, the reason why the Eocene is considered as the upper limit time is because there is an uncertain issue of oil cracking rate. That is to say how long it takes for a certain scale paleo-oil pool to fully crack into gas, after the oil reservoir entered into oil cracking temperature window, since in different geological environments and media, the oil cracking rate is different. At a
great cracking rate, it is possible that the oil pool was fully cracked into gas, when its temperature was not reached 210 °C. Therefore, the time space determined in this paper is actually from the starting time to the upper limit time.

CONCLUSIONS

Oil cracking history can be reconstructed by the oil cracking temperature window method. Taking the oil reservoir as a case study object, based on burial and paleo-heat flow history reconstructions, we firstly reconstructed the reservoir temperature history and then combined the reservoir temperature history with oil cracking temperature window to determine the oil cracking time.

Paleo-geothermal gradient inversion revealed that Weiyuan area has undergone a large scale of erosion in Himalayan period. The erosion thickness reached 2889 m. Weiyuan area experienced heat flow elevating in Paleozoic-early Permian and decreasing from the late Permian to the present day; the peak of heat flow (85.3 mW/m²) was reached at the end of the early Permian (258 Ma bp). The present-day surface heat flow is 65 mW/m².

The present-day Weiyuan Sinian gas field origins from a paleo-oil pool which was cracked into gas at high temperature. The oil cracking into gas temperature window is roughly between 160 to 210 °C. By using the oil cracking temperature window method, it is deduced that the process of oil cracking into gas in Weiyuan paleo-oil pool started at the beginning of the late Triassic and ended in the Eocene (upper limit time).

ACKNOWLEDGEMENTS

This study is financially supported by the National Key Foundational Research and Development Project (Grant No: 2012CB214806) and the National Science & Technology Special Project (2016ZX05061001003). Special thanks are given to the reviewers for their useful comments and suggestions.

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