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**Phase States of Hydrocarbons in Chinese Marine Carbonate Strata and Controlling**  
**Factors of Their Formation**

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Chinese marine carbonate strata were mainly deposited before the Mesozoic. Because of this long burial time, they have experienced multiple phases of thermal and tectonic events, which in turn have brought multiple periods of hydrocarbon generation, secondary oil cracking, remigration and re-accumulation in reworked structures. Phase states of Hydrocarbons are rather different among typical Chinese marine basins (i.e. Tarim, Sichuan and Ordos Basins). For Sichuan and Ordos Basins, hydrocarbons was phase-stated as natural gas, with burial depth varying from 200 m (204.5m for gas producer in four member of lower Triassic Jialingjiang FM in Weiqian1 well) to 6590 m (6593m for Upper Permian Changxing gas layer in Yuanba-2 well), while both liquid and gas phase state exists in Tarim Basin. In an extreme case, the Cambrian discovery of 8400 m deep by Tashen1 well still contains liquid phase hydrocarbons. This raises the question whether the different characteristics of source rocks or the different basin thermal histories has caused this fluid phase difference. This research casts a light on the controlling factor of hydrocarbon fluid phase by discussing the processes for hydrocarbon generation and alteration in these three basins, incorporating understandings of kerogen types, hydrocarbon precursors, basin thermal histories, source rock maturity, burial histories and hydrocarbon generation simulations in these basins. The discussion is further constrained by results from regional structure evolution and gas geochemistry.

1. Difference in basin thermal histories of the Chinese sedimentary basins

The thermal histories are different among different Chinese marine basins. The present geo-thermal flow of Tarim Basin is 35-75mW/m<sup>2</sup>, with an average value of 43.5mW/m<sup>2</sup>. The reconstruction of paleo-heat flow and vitrinite reflectance (Ro) in the Tarim Basin indicates a gradual decline of heat flow in geological history: the Cambrian 65-75mW/m<sup>2</sup>, the Late Silurian 60-66mW/m<sup>2</sup>, the Late Permian 55-60mW/m<sup>2</sup>, the Late Cretaceous 50-55mW/m<sup>2</sup>. Thus, the heat flow in the Tarim basin has gradually declined after the Paleozoic, although the exact thermal histories are somewhat different between the eastern, middle and western part of the basin. The current thermal gradient is 22-25°C/km.

The concurrent geo-thermal flow of Sichuan Basin is 35.4-68.8mW/m<sup>2</sup>, averaging

53.2mW/m<sup>2</sup>. The basin has experienced the Late Paleozoic to the Early Permian heat flow increase followed by a decrease period from the Late Permian till the present. In the Early Permian the heat flow had reached the maximum value of 71-77mW/m<sup>2</sup>, and then declined since the Late Permian. Through the Mesozoic and Cenozoic, different structure elements in the Sichuan Basin have different decline rates of heat flow. This has led to heat flow differences across the basin, with Middle and Southern Sichuan basin maintaining the highest thermal gradient, locally reaching 32°C/km or higher.

Ordos Basin shows a modern geo-thermal flow of 43-70mW/m<sup>2</sup>, averaging 60mW/m<sup>2</sup>. During Paleozoic and Early Mesozoic, the paleo-thermal gradient is relatively low, with 22-25°C/km (45-55mW/m<sup>2</sup>). But strong tectonic activity of Late Mesozoic (Yanshan Movement) has significantly increased the paleo-thermal gradient, reaching 33-48°C/km (70-80mW/m<sup>2</sup>). After Early Cretaceous, the entire basin uplifted and subjected to erosion, causing decline in geo-thermal gradient and dropping of formation temperature.

## 2. Difference in hydrocarbon generation history for marine source rocks in the Chinese basins

The pre-Mesozoic source rocks of the Chinese marine basins were mainly deposited in the passive margin slope and ramp settings, or intra-cratonic basins, under a general extensional tectonic environment. These source rocks are mainly dark shale or calcareous mud, but locally coal measures of marine-continent transitional facies also exist, such as Upper Permian Longtan Formation in Sichuan Basin and Carboniferous-Permian coals in Ordos Basin.

According to geochemical studies on outcrop and core samples, the main marine source rocks of Tarim Basin were deposited in the Early to Middle Cambrian and the Middle to Late Ordovician, with TOC value of 0.5%-5.54%, and kerogen type is I-II<sub>1</sub>, hydrocarbon precursor is composed of dinophyceae and planktonic algae. Middle and Lower Cambrian source rocks has the lowest thermal maturity in Bachu Uplift, with equivalent vitrinite reflectance (VR<sub>o</sub>) of 1.44-2.06%, and the highest maturity in Manjiaer Sag, VR<sub>o</sub> up to 4.0%. The Middle and Upper Ordovician source rocks have reached mature in the Tazhong and Tabei Uplifts, with VR<sub>o</sub> between 0.81-1.31%, and no obvious correlation between VR<sub>o</sub> and depth is observed. However, this source rock has reached high to over mature stage in Tadong Uplift, and average VR<sub>o</sub> of 2.06 in TD1 Well.

Sichuan Basin's marine source rocks were deposited during the Early Cambrian, Early Silurian and Late Permian. TOC varies between 0.5-8.0%, kerogen type is I-II<sub>1</sub>, hydrocarbon precursor is plankton algae and benthic algae. Lower Cambrian and Silurian source rocks are mostly high to over mature, with VR<sub>o</sub> between 1.77-3.38%. The maturity of Upper Permian source rock varies significantly across the basin, in the Guanyuan-Wanyuan-Chengkou region, which is the tectonic front of Longmen-Micang-Daba uplifts, the source has only reached early mature – mature stage, but in other regions the source rock has reached over mature stage, such as the Upper Permian source rock in Northeast Sichuan Basin has reached a VR<sub>o</sub> value of about 3.94% in Puguang-5 Well. The geochemical parameters (i.e. correlation between hopane/sterane and tricyclic terpane/hopane, aromatic compound series, and stable carbon compositions of saturated hydrocarbon and aromatics) have shown that the reservoir asphalts are closely related to the Upper Permian source rock, while unrelated to Lower Permian and Silurian source rocks.

Model simulation of burial and hydrocarbon generation histories of Puguang Field indicates that the hydrocarbon charging period would be about 200-160 Ma, matching with the main hydrocarbon generation period of the Upper Permian source rock, while different from the main period of the Silurian source rock (main period >235 Ma). Thus, the natural gas in the marine strata of Northeast Sichuan (including Puguang Field) is mostly sourced from the Upper Permian source rocks rather than the Silurian or Lower Permian source rocks.

The Middle-Lower Ordovician marine source rocks of Ordos Basin are mainly distributed in the West and Southwest part of the basin, while absent in the centre of the basin. The special distribution of the source resembles an L-shape. TOC varies between 0.22-3.3%, averaging 0.93%, and the main kerogen type is II<sub>1</sub>, hydrocarbon precursor consists of acritarchs, algae and lower eukaryote. The present VR<sub>o</sub> value for marine source rock is usually more than 2.0%, in high to over mature stage.

In the correlation between  $\delta^{13}\text{C}_1$ - $\delta^{13}\text{C}_2$  and  $\delta^{13}\text{C}_2$ ,  $\delta^{13}\text{C}_2$  values in natural gas of the Northeast Sichuan is less than -28‰, and is negatively correlated to the  $\delta^{13}\text{C}_1$ - $\delta^{13}\text{C}_2$ . These features are similar to those in Jinbian Gas Field in Ordos Basin and Hetianhe Gas Field in Tarim Basin, natural gas is of oil-type gas with more negative  $\delta^{13}\text{C}$  ( $\delta^{13}\text{C}_2 < -28\text{‰}$ ). However, natural gas in the Guangan and Chongxi Gas Fields in the Middle Sichuan and Kela2 Gas Field in Tarim Basin is typical coal-type gas, with less negative  $\delta^{13}\text{C}$  ( $\delta^{13}\text{C}_2 > -28\text{‰}$ ).  $\delta\text{D}_1$  values more than -140‰ indicate the source rock deposition under marine environment. A modified model of C<sub>2</sub>/C<sub>3</sub> and  $\delta^{13}\text{C}_2$ - $\delta^{13}\text{C}_3$  pattern is applied to distinguish between primary cracking gas and secondary cracking of oil and gas, the results shows that all natural gas in Sichuan and Ordos Basins was fallen into oil and/or gas secondary cracking, while the gas in Tarim Basin contain both primary cracking gas and secondary cracking of oil and gas. The portion of oil-cracked gas depends on the preservation condition of oil, and the paleo-uplift is favorable for oil existence.

### 3. Difference in hydrocarbon accumulation processes in Chinese marine basins

Hydrocarbon accumulations in Chinese marine strata are characterized by multiple source rocks and multiple phases of generation, with several periods of charging and migration, accompanied by secondary cracking and other fluid-gas phase changes, and the accumulations are subjected to readjustment later.

Tarim Basin as a typical superimposed basin has complicated thermal and burial histories. The marine source rock has experienced multiple stages and phases of hydrocarbon generation. The oil was formed from the Middle-Lower Cambrian source rock and accumulated in the early-middle Ordovician period. But the strata uplift has tempered the hydrocarbon generation and part of accumulations was destroyed during Silurian-Devonian period. The oil generation was resumed during the Carboniferous but maturity of the source rock evolved little, staying in the oil and condensate generation phase for a long period of time. The maturity evolution of source rock accelerated during the Cretaceous, and early condensate was cracked into dry gas. Some deep buried oil was also cracked into gas. The Middle-Upper Ordovician source rock entered oil generation window in the Carboniferous, and became mature in the Cretaceous, generating mainly oil and condensates, and still in this stage at the present time.

Sichuan Basin also shows multiple phases and stages of hydrocarbon generation. The

Cambrian and Silurian source rock of the basin entered the peak generation phase before Triassic, with correspondent accumulations formed in suitable paleo-traps during the same period. But during the Middle Jurassic (Yanshan Movement), the oil in the early accumulation had cracked into gas, while the Permian source rocks had entered the peak generation phase, and charged reefal and lagoonal reservoirs. These hydrocarbons (oil and condensate) had cracked into gas during the late stage of Yanshan Movement. During the Himalaya Movement, the structural trap formation and reworking of earlier structures has caused the readjustment and re-accumulation of cracked natural gas.

The two settings of marine source rocks in Ordos Basin had become mature in the Late Triassic, and oil was accumulated at the slope zone of the central paleo-uplift. During Jurassic to the Early Cretaceous, the increasing burial depth and rising geo-thermal gradient has caused the secondary cracking of oil. The large scale uplift and compression in the eastern part of the basin during the Late Cretaceous had caused the gas migration toward the eastern basin and present-day accumulation as Jinbian Gas Field. Where absence of Carboniferous Fe bauxitite cap rocks at top of Ordovician paleo-weathering crust has caused the coal-type gas sourced from Carboniferous-Permian coals to migrate along the unconformity, and to mix with local oil-type gas.

#### 4. Conclusions

Although the Chinese marine carbonate strata has multiple source rocks and multiple phases of hydrocarbon generation stages, the basin thermal history and source rock maturity is the main controlling factor of hydrocarbon phases. The kerogen type and hydrocarbon precursor imposes relative small influence. Because Tarim Basin has experienced continuous decline of heat flow since the Paleozoic, oil in structural high could be preserved, and thus both oil and gas can be discovered in the basin. Sichuan Basin was also subjected to the decline of heat flow since Permian, but the overall heat flow remained to be high. Ordos was previously a “cold basin”, but the thermal event during the Cretaceous has caused the high maturity of the source rocks. Thus, the liquid phase hydrocarbons are difficult to preserve in the Sichuan and Ordos Basins, and mostly cracked into gas, in contrast to that of Tarim Basin.