Two typical accumulation mechanisms of marine natural gas reservoirs in China: Case comparison between Tarim Basin and Sichuan Basin

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Abstract. Marine basin contains abundant natural gas resources in China, especially the Palaeozoic is becoming a hotspot of natural gas exploration, wherea series of large gas fields have been constantly discovered. It is broad consensus that high-post mature source rocks (SRs) is gas-prone and thermal sulphatereduction (TSR) could accelerate the hydrocarbon cracking, which would be the dominant origin of gas enriched in the Palaeozoic of petroliferous Basin in China. In addition, these cracking gas is rich in H2S and CO2, due to the marine source rocks are rich in argillaceous sedimentary and sulfates. Nevertheless, the accumulations of gas reservoirs were obviously different between two giant marine craton in China. Sichuan basin is abundance of marine cracking gas, which was mainly generated around 257-205Ma. But it is rarely found marine oil reservoirs in Sichuan basin. In contrast, abundance of marine condensates are rich in Tarim Basin. Gas washing and migration fractionation effect would be the major formation mechanism of secondary condensates, which coexist with dry gas and normal oil reservoirs. These various reservoirs orderly distribute around source rocks in the Manjia’er sag of Tarim Basin. The origin of marine gases was mainly mixed from the hydrocarbon cracking derived via TSR and the different stages of thermal evolution of marine SRs, as well as portion of humic gas. Therefore, the carbon isotopic composition of diverse gas reservoirs is quite complex and the phenomenon of isotopic reversal $\delta^{13}C_1 > \delta^{13}C_2$ is pervasive. Eventually, two typical accumulation mechanismsof marine natural gas reservoirshave been established. It could be inferred the exploration of cracking gas is given priority in Sichuan Basin and marine secondary condensates should be paid more attention during the petroleum exploration in Tarim Basin.

1. Introduction

The geological theory and exploration techniques of natural gases have made great progress since 1983 in China. A group of large and medium gas fieldshave been successively found in the Sichuan, Tarim, Junggar and Qaidam Basin, especially in the deep of Songliao Basin. The natural gas exploration had proceeded into a rapid development period in China. In recent years, with the accelerationof deep exploration, a series of gas exploration breakthrough were achieved in the Palaeozoic of Sichuan, Ordos, and Tarim Basin. Especially, Anyue gas field reached nearly trillion reserve, as well as Sinian gas reservoirs in the central Sichuan Basin and Yuanba gas reservoir.
Furthermore, large marine gas fields had been discovered in the Ordovician of Tarim basin, involving Tazhong, Lugudong and Hetianhe gas reservoirs. All of these exploration progress exhibit giant potential of marine gas resource. Associated with the significant progress of Palaeozoic marine petroleum exploration, some new issues are raising in front of geologists, such as why gas reservoirs are abundant in Sichuan Basin, while condensate reservoirs are rich in Tarim basin. Traditional indexes tracing origin and genesis, such as composition and carbon isotope of natural gas, encounter new difficulties due to complex accumulation process. Therefore, it would be necessary to clarify the complicated formation mechanism of giant marine gas reservoirs, which could be benefit to continuously safeguard deep marine exploration access to new discoveries.

2. Geochemical characteristic of giant gas reservoirs in the Sichuan Basin and Tarim Basin

Through the comparisons of composition and isotopic characteristics between the giant gas reservoirs in the Sichuan Basin and Tarim Basin, the former is major feature of high pure gas with dry coefficient greater than 0.99, while component content of C1+ less than 0.5%; the latter is abundant of condensates, gas reservoirs only accumulate in Hetahe area located in the southwest Tarim Basin (Table 1). The type of source rocks, stratigraphic lithology, thermal evolution and tectonic cycle history could be the major impact factors on the differences between physicochemical properties of gas reservoirs in the Sichuan Basin and Tarim Basin.

| Basins   | Gas Fields  | Reservoirs | Proved reserves ($\times 10^8 m^3$) | CH4 (%) | C1/C1+ | Non-hydrocarbons | $\delta^{13}C_1$ (%) | $\delta^{13}C_2$ (%) | Source type    |
|----------|-------------|------------|------------------------------------|---------|--------|------------------|-----------------------|---------------------|----------------|
| Sichuan  | Puguang     | T1f, P2    | 2510.7                             | 76.42   | 0.999  | 23.21            | -30.47                | -27.4               | Coal, Marine shale |
|          | Luojizhai   | T1f        | 581.08                             | 81.6    | 0.999  | 18.18            | -30.38                | -29.4               | Marine shale     |
|          | Wubaiti     | C2, P2     | 409                                | 96.6    | 0.991  | 2.53             | -31.8                 | -35.51              | Marine shale     |
|          | Weiyuan     | P1, Z2     | 408.61                             | 86.58   | 0.999  | 12.9             | -32.5                 | -31.95              | Marine shale     |
|          | Shapingchang| C2         | 397.71                             | 95.17   | 0.995  | 3.02             | -32.31                | -36.12              | Marine shale     |
|          | Wolonghe    | C, P, T    | 380.52                             | 91.82   | 0.995  | 3.14             | -32.84                | -35.59              | Marine shale     |
| Tarim    | Eastern     | O2-3       | 499.51                             | 91.71   | 0.992  | 4.1              | -34.8                 | -37.5               | Marine shale     |
|          | Lungu       | O1         | 2821.48                            | 86.05   | 0.971  | 10.5             | -54.4                 | -38.2               | Marine shale     |
|          | Tazhong I   | O2-3       | 972.61                             | 82.35   | 0.961  | 14               | -38.5                 | -36.6               | Marine shale     |
|          | Hetianhe    | C          | 616.94                             | 78.72   | 0.965  | 18               | -37.2                 | -36.48              | Marine shale     |

3. Comparison of geochemical characteristic between the dominant marine source rocks in the Sichuan Basin and Tarim Basin

The major source rocks (SRs) were developed in the Cambrian, Ordovician and Silurian of the two marine basins, and there are obvious differences in thermal evolution extent between them (Table 2). Cambrian SRs are widely distributed in Tarim basin and Sichuan basin, and the thermal maturity reached high-post stage ($R_o>3.5\%$) (Table 2). It has been confirmed that Cambrian is the main SR of Anyue giant gas field. Maturity of Cambrian SR was mostly more than 2% in Tarim basin, which was thought to be the main SR of Hetianhe gas field. The Ordovician SR are mainly developed in Tarim Basin, with moderate thermal evolution and “oil window” maturity, which is an important reason for marine oil rich in Tarim Basin. The Ordovician SR in Sichuan Basin continuously deposited below the lower Silurian, which are widely distributed in the eastern Sichuan Basin and become the main producing strata of shale gas in the eastern Sichuan area. These three sets of typical marine source rocks are mainly gas-prone. Except for the low maturity of Ordovician source rock in Tarim Basin, the other two sets of source rocks have evergenerated large amount of ancient oil reservoirs, but oil cracked and turned into gas in the later stage.
4. Formation and accumulation of marine gases
4.1. Accumulation and reservoir evolution of marine gases in the Sichuan Basin

In the northeast Sichuan, Feixianguan gas reservoirs were rich in H2S, while carbon isotopic range of gases was not various. The $\delta^{13}C_1$ (Methane) was between -29.52‰ ~ -31.5‰, and the $\delta^{13}C_2$ ranges from -29.4 ‰ to -32.39 ‰, both of which were slight heavier 1‰ ~ 2‰ than that of feixianguan gas reservoirs in other areas. This is derived from the result of TSR reaction, but not the difference of gas sources. A similar situation was also reported in the Hetianhe gas field of Tarim\cite{1}, the carbon isotope of gases with high H2S content was slight heavier 1‰ to 3‰ than that of gases with low H2S content. The $\delta^{13}C_2$ possessed a good inheritance of parent material, which of oolite beach gas reservoirs was -28 ‰, indicated sapropel. In addition, it was discovered $\delta^{13}C_1$ is -29.83 ‰, the $\delta^{13}C_2$ is -32.39 ‰, reverse of $\delta^{13}C_1$ and $\delta^{13}C_2$ occurred in the feixianguan reservoir of well- Dukouhe 4 in eastern Sichuan Basin. The similar characteristics was also existed in Carboniferous and Permian gases (Figure 1). Therefore, the TSR reaction might be the important cause of isotope reversal between methane and ethane, as well as hydrocarbon cracking.

![Figure 1](image-url)

Figure 1. The relationship diagrams between relative amount and isotopic characteristics of CH4 and C2H6 in the Sichuan basin.

(1) The thermal evolution of Lower Cambrian source rock and accumulation of natural gases. Lower Cambriansource rocks mainly distribute in the south Sichuan Basin, which reached the dry-gas generation stage in the late Hercynian-early Indosinian (250-205Ma). However, the petroleum trap in Feixianguan formation of Triassic had not yet formed. Therefore, the Lower Cambrian SR might not contribute to gas reservoirs in the Feixianguan formation of Triassic.

(2) The evolution of Silurian SRs and accumulation of natural gas reservoirs. Silurian Longmaxi formation got into oil-generation peak in the late Triassic. The evolution history of Silurian SRs consisted with the formation period of petroleum trap in the Feixianguan formation of Triassic.

In the middle of the Yanshan period (~100Ma), ancient oil reservoirs began to crack along with the basin subsidence (Figure 2). The hydrocarbons in the Silurian reservoirs initialed to crack into gas with high content of H2S, which resulted in the formation cracking-gas reservoirs in Triassic.

(3) Thermal evolution of source rocks in Permian and accumulation of gas reservoirs. Maturity of Longtan formation SRs has reached 1.9 ~ 2.9%, which has got into high mature stage. And these
Permian SRs reached oil-generation peak during the period of late Jurassic - early Cretaceous, which consisted with the adjustment and finalization period of gas reservoirs in the Feixianguan formation of Triassic (Figure 2). Therefore, Longtan Formation SRs should be is one of the main origin of Feixianguan gas reservoirs.

4.2. Accumulation and reservoir evolution of marine gases in the Tarim Basin

In the marine strata of Tarim Basin, the large gas reservoir (Hetianhe gas field) and eastern Lungu condensate have been discovered in succession. Recently, a number of new discoveries have been achieved during the strengthen exploration around the Tazhong I slope zone. Contrast to the Sichuan basin, marine gases in Tarim basin were mostly characterized by condensate reservoirs, the large marine pure gas reservoirs were only found in the Carboniferous of Hetianhe area. And, the dry coefficient of marine gas reservoirs in the Sichuan basin accumulation was abnormally high (mostly dry coefficient greater than 0.99), moisture components of gas was generally less than 0.5% (Table 1). Marine gas reservoirs of the Tarim basin were mainly distributed in northern and central, two inherited uplift. Through the comparison of isotopic characteristics, it could be demonstrated that carbon isotopic is positive sequence arrangement with the carbon number in the center of Lunnan area (Fig. 3), which conforms to the normal kerogen evolution characteristics of hydrocarbon generation. However, the carbon isotopic of gases in the western Lunnan area lighter than -41‰, which indicated the origin of biodegradation (Fig. 3). Especially in eastern Lungu area, the dry coefficients of gases were 0.98 greater than those of any other area of Lunnan uplift. The isotopic composition of gases is antitone sequence arrangement with the carbon number, and the distribution of carbon isotope value is consistent, δ¹³C₁ > δ¹³C₂ > δ¹³C₃ (Fig. 3), carbon isotope of methane form -36‰ to -34‰, carbon isotope of ethane form -37.5‰ to -34.5‰, and carbon isotope of propane form -30.2‰ to -34.7‰. It was showed that the gas of condensates is likely to be invaded with high mature oil-cracking gas. And, the gases in the reservoirs of overlying Carboniferous and Triassic are also appearance of the differences between the eastern and western. However, according to the result of oil-source correlation, the oil-source of three sets of main oil-bearing strata is mainly consistent[2]. Therefore, the diversity of petroleum properties in different areas is likely caused by the various secondary changing after the initial accumulation.
Figure 3. The relationship diagrams between relative amount and isotopic characteristics of CH₄ and C₂H₆ in the north Tarim basin.

Two sets of marine source rocks mainly developed in the Paleozoic group of Tarim Basin, which had experienced multi-phase hydrocarbon generation and accumulation, multi-period adjustment and transformation in the long process of geologic evolution history. It was indicated that the Caledonian to early Hercynian (~360 Ma), the late Hercynian (~250 Ma) and late Himalayan (~20 Ma) were the three key geological periods for the formation of such a complex petroleum system, based on the comprehensive research of basin evolution (Fig.4), hydrocarbon generation and the history of accumulation and adjustment[3]. The first two periods is given priority to oil accumulation, but the late Himalayan is given priority to gas charging. And the ancient reservoirs have experienced the adjustment, renovation and destruction in the late Himalayan.

Figure 4. Evolution history of stratigraphic settlement in the north Tarim Basin.
Especially since 20 Ma ago, thick Cenozoic covered on the sedimentary combination of Paleozoic and Mesozoic, so that the evolution of marine source rocks and the generation of hydrocarbons had been accelerated. Cambrian hydrocarbon source rocks in the Manjia’er sag entered over-mature phase, the gases generated from oil-cracking, migrated and charged from east to west. The high-yield gas-wells are mainly distributed in the eastern region, along the deep and large fracture. It was indicated that the late high-maturity gases came from the deep. This stage of the gas charging washed early oil reservoir, which brought about the formation of secondary condensates reservoir. In the northern Tarim, the condensates of eastern lungu originated from the gas washing in the late Himalayan. The oil-cracking gas was much dry. when them charged and intruded into ancient reservoirs, one side displace the normal oil in the reservoir, these oil migrated upward and accumulated in the overlying stratigraphic trap along faults[4], ether effused and accumulated in the upward direction of reservoir along carrier bed. On the other hand, the charging dry gases dissolved weight-light portion of oil, which led to form second condensates. The geochemical characteristics of the second condensates were impacted by the tectonic movement in the late Himalayan.

5. Conclusions
Both of Tarim Basin and Sichuan Basin belong to marine craton basin, but two typical accumulation models of marine gas are finally formed in China due to the difference of tectonic evolution cycles between two basins. Three sets of marine SRs were developed in Sichuan Basin, which experienced hydrocarbon crackingin the deep strataaround 175Ma, and gas reservoirsexhibited the characteristics of continuous accumulation. Two sets of Cambrian and Ordovician SRs were developed in Tarim Basin, which respectively evolved into the oil-generation peak around 360Ma and 250Ma. The geological process of deepest settlement occurred around 20Ma, abundant condensates in Tarim Basin have been confirmed to be derived from mixture of later cracking gas with early oil reservoirs. Therefore, Sichuan Basin and Tarim Basin separately represent two typical accumulation models of marine gas reservoirs. In Sichuan Basin, the gas resources predominantly derived from TSR and hydrocarbon cracking, and exhibited the characteristics of the continuous accumulation. Otherwise, the secondary condensates in Tarim Basin almost originated from the migration fractionation effect of hydrocarbon cracking and early oil reservoirs.

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