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Original article

Hydrocarbon origin and reservoir forming model research of Longwangmiao Formation, Moxi-Gaoshiti area, Sichuan Basin

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A R T I C L E I N F O

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ABSTRACT

This paper focuses on the Longwangmiao gas reservoir in Moxi-Gaoshiti area, Sichuan Basin. Starting from the tectonic evolution perspective, though comparing biological marker compound and analyzing fluid inclusions, the oil & gas origin and accumulation evolution of Longwangmiao Formation are systematic studied with reference to the burial-thermal evolution of single well geological history in the study area. It is suggested that the oil & gas reservoir is generally characterized by early accumulation, multi-stage filling, late cracking and later adjustment. The oil and gas were mainly sourced from lower Cambrian Qiongzhusi Formation, partly from the Permian source rock. During the geological period, 3 major oil & gas fillings occurred in the Longwangmiao Formation, namely Caledonian-Hercynian filling that was small in scale and produced the first phase of paleo-oil reservoir that soon destroyed by Caledonian movement uplift, large-scale Permian filling that gave rise to the second-phase of paleo-oil reservoir and the Triassic-Jurassic filling that enriched the second phase of paleo-oil reservoir. Finally, the paleo-oil reservoir experienced an in-situ cracking during the cretaceous period that gave rise to a natural gas reservoir and left behind carbonaceous bitumen and oily bitumen in the holes of the Longwangmiao Formation.

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1. Introduction

The Longwangmiao Formation in Moxi-Gaoshiti area is one of the hotspot in domestic natural gas exploration. In 2014, the biggest single mono-block marine-facies carbonate gas field so far was found in the Longwangmiao Formation, with a proven reserve of 4403.85 \times 108 m³, which reveals the great exploration potential of the high-evolution natural gas reservoir in Sichuan Basin. Studies show that the Moxi-Gaoshiti area has favorable

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conditions for petroleum geology, with massive Sinian – Quaternary depositional thickness, multiple sets of high-quality hydrocarbon source rocks and broad exploration field [1]. The current research has mainly focused on the controlling factors and depositional model of the reservoir [2-5], rather than the accumulation mechanism of Longwangmiao Formation. However, knowledge of this is the key to further high-efficient exploration of oil & gas reservoirs [6,7]. For this reason, this essay, based on the sufficient investigation of preliminary exploration and research results, start from the source of bitumen in Longwangmiao Formation, combined with the analysis of fluid inclusion composition and structural evolution, conducted the reconstruction of the accumulation process, and systematically studied the characteristics of oil and gas enrichment and accumulation, in hope of providing more scientific basis to expand the exploration of the Longwangmiao Formation in Sichuan Basin.

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Fig. 1. Location, structure and stratigraphic column of the study area.

2. Geological background

The Moxi-Gaoshiti area, located in the Suining city of Sichuan province, Anyue County of Ziyang city and Tongnan county of Chongqing city, tectonically, is a part of the Leshan-Longnvsi paleouplift in the central west of the medium slant gentle belt on the paleouplift in central Sichuan (Fig. 1), with a total area of 2330 km². Controlled by the paleo-topography of the paleouplift [8], The Longwangmiao Formation in the study area is mainly of granular carbonate deposits and is in conformable contact with the overlying Gaotai formation of mixed tidal flat deposits and the underlying Canglangpu Formation of shelf clastic deposits in the undenudation area [9] (Fig. 1).

Studies of structural evolution show that [10-13] Leshan-Longnvsi palaeohigh is a uplift controlled by basement and fracture under tensile stress with some succession. It began to take shape after a syndepositional uplift and denudation uplift during Sinian Dengying period, which is called "low uplift". Then, it developed rapidly in late Lower Cambrian when the basement rift activities were the most active, which is called "high uplift". The Caledonian movement in late Silurian period caused Sichuan Basin to lift greatly, and also greatly denuded the Cambrian -Silurian strata in middle Sichuan, and caused the Longwangmiao Formation in local area to be exposed or almost exposed and caused the porous and permeable reservoir preserved by retention processes to be transformed by weathering karstification. As a result, the quality reservoir in Longwangmiao Formation was formed [2]. Subsequently, It took deposition again and consequently the Longwangmiao Formation are in unconformable contact with the overlying strata of lower Permian in the denuded zone. The range of Leshan-Longnvsi palaeohigh were basically established and the macro tectonic features were formed during the Caledonian period, which provided conditions for successive development in later structural event. After this period, the palaeohigh entered developmental and evolutionary stage and successively experienced Hercynian, Indo-Chinese, Yanshanian and Himalayan evolutionary stage. In the latter three stages, the palaeohigh developed successively but also experienced adjustment, transformation and destruction.

3. Comparative analysis of bitumen and hydrocarbon sources of Longwangmiao reservoir

Bitumen, as a product in the high evolutionary stage of oil, is closely related to oil and gas in terms of genesis. Part of the bitumen was formed in a process where crude oil from source rocks accumulated in the reservoir, then went through biodegradation, oxidation and water-washing. The bitumen could eventually generate natural gas by high temperature pyrolyzing. Therefore, bitumen is of special significance as to indicating the genesis of relevant oil & gas and effective source rock. Another part of the bitumen was formed in the process where crude oil cracked into gaseous hydrocarbon (Wet gas) and young bitumen in semi-solid state with high plasticity [14]. This shows bitumen, regardless of the genesis, is all excellent for the study of hydrocarbon origin of natural gas reservoir [15].

95 samples were selected in the study. The microscopic observation of the bitumen shows that mainly two types of bitumen were found in the Longwangmiao Formation in Gaositi-Moxi area – carbonaceous bitumen (Fig. 2a) and oily bitumen (Fig. 2b). Both of them were found in the dissolved pores of dolomite, they can exist on their own or mixed together. The carbonaceous bitumen has higher maturation and evolution stage, does not respond to fluorescence and is widely distributed in the reservoir while oily bitumen is still in maturation stage, responses well to fluorescence and is less distributed (Fig. 2c). The co-existence of carbonaceous bitumen and oily bitumen may indicate that the bitumen source of the Longwangmiao Formation is relatively complicated and also may indicate the complication of the origin of source rocks.

Research has already shown that the source rocks of Longwangmiao Formation in the Moxi-Gaoshiti area are mud shales from the underlying Qiongzhusi Formation [3,16,17]. The comparison of biological marker compounds between Longwangmiao bitumen and Qiongzhusi mud shales shows that the distribution characteristics of bitumen terpane and gonane are significantly similar to that of Qiongzhusi mud shales (Fig. 3). In terms of carbon isotopes composition of Longwangmiao bitumen and source rocks (Fig. 4), it is also suggested that the bitumen



Fig. 2. Characteristics of bitumen in the Longwangmiao Formation, Moxi-Gaoshiti area. (a), (b)Well Moxi 12, 4632.61 m, non-fluorescing carbonaceous bitumen, (a) plane polarized light, (b) fluorescent light; (c), (d) Well Moxi 12, 4655.22 m, fluorescing oily bitumen, (c) plane polarized light, (d) fluorescent light; (e), (f) Well Moxi 12, 4655.22 m, co-existence of carbonaceous bitumen and oily bitumen, (e) plane polarized light, (f) fluorescent light:



Distribution characteristics of gonane

Fig. 3. Distribution characteristics of biological marker compounds.

Sample		Carbon isotope characteristic (%)
Natural gas	Longwangmiao Format- ion in Moxi-Gaoshiti area	• • • • • • • • CH_4 • • • • • • • • C_2H_6
	Longwangmiao Format- ion of Well Baolong 1	◆ CH4 ◆C2H6
Bitumen	Bitumen from Longwang- miao Formation	Chloroform asphalt"A" ◇ ◇ ◇ ◇ ◇ ◇ ◇ ◇ ◇
Source rock	Mudstone from Qiongzhu- si Formation	•••• • ••• Kerogen Chloroform asphalt"A"∞∞∞ ∞ ◊
	Mudstone from3rd member of Dengying Formation	◆ ◆ ◆ Kerogen Chloroform asphalt"A" ◊ ∞ ◊
	Source rock from Permian	Kerogen Chloroform asphalt"A"

Fig. 4. Carbon isotope characteristics of natural gas, bitumen of Longwangmiao Formation and source rocks.

source of Longwangmiao Formation in Moxi-Gaoshiti area is influenced by Qiongzhusi Formation. Because hydrocarbon generated early in Qiongzhusi Formation, as it buried deeper, the liquid hydrocarbons filling Longwangmiao Formation became more mature until they became over-matured and left carbonaceous bitumen behind.

As mentioned above, aside from carbonaceous bitumen, there is also less matured oily bitumen in Longwangmiao reservoirs. Previous research has shown that the co-existence of two types of bitumen is due to fillings from different source rocks in different periods [18], or fillings from the same source rock in different periods. As for the hydrocarbon filling in the Longwangmiao Formation in Moxi-Gaoshiti area, the case is different. After the Caledonian movement, the oil generation of Qiongzhusi source rocks reached its peak during Permian-Middle Triassic, and they became over matured in late Triassic, eventually stopped generating hydrocarbon by entering into deep burial stage [17]. So the possibility that the source rock of Qiongzhusi Formation filled in multiple phases is ruled out, and the hydrocarbon filling that gave rise to the oily bitumen of Longwangmiao Formation must be later than the hydrocarbon filling of Qiongzhusi Formation. With reference to the distribution layer of the source rocks in Moxi-Gaoshiti area, the study area shows an absence of Silurian source rocks and this means the most possible hydrocarbon origin should be the overlying Permian source rocks. In terms of the distribution characteristics of bitumen n-alkanes in the Longwangmiao reservoir, it is quite similar with Permian source rocks, but significantly different from Qiongzhusi Formation source rocks (Fig. 5), which indicates that the origin of Longwangmiao bitumen was influenced by Permian source rocks.

4. Oil and gas accumulation phases

In this study, 60 points were selected in Well Moxi 12 and 13 for homogenization temperature test on inclusions. The inclusions are mainly found in the host dolomite minerals. They are mainly two-phase water-salt inclusions which are usually superimposed on bitumen inclusions (Fig. 6). Under microscope, the inclusions are mainly oval with the size ranging from 5 to 10 um. Their bubble diameters are small, with most of them around 2um. The bar chart of salt-water inclusions' homogenization temperature shows three phases of fluid filling, namely 110-130 °C filling, which is possibly related to Caledonian -Hercynian fluid fillings, 130–160 °C filling, which is possibly related to Indo-Chinese - Yanshanian filling, and 160-240 °C filling, which is possibly related to Yanshanian filling. With reference to the burial-thermal evolution of single well source rocks geological history (Fig. 8), there were mainly two sets of source rocks in the depositional-burial process and accordingly, there were three phases of fillings. They are, respectively, early filling from Qiongzhusi mature source rocks that mainly happened during middle Cambrian-Silurian period, with small charge volume, weak influence and the corresponding inclusions are 110~130 °C; middle filling from Qiongzhusi burial source rocks that happened mainly in Permian-Triassic period and caused by the reburial of basin in Permian, with large charge volume, strong influence and the corresponding inclusions are 130 ~ 160 °C; and the later filling from Permian mature source rocks that happened mainly in Triassic-Jurassic, with small charge volume, medium influence and the corresponding inclusions are 160 ~ 240 °C. The last filling is responsive to the above-mentioned organic geochemistry characteristics of bitumen and mainly contributed to the oily bitumen in the Longwangmiao Formation. Fluorometric analysis on abundant slices shows that carbonaceous bitumen accounts for about 80% while oily bitumen accounts for about 20%. This indicates that lower Cambrian Qiongzhusi Formation was the major hydrocarbon source while the contribution from Permian source rocks was insignificant (see Fig. 7).

5. Accumulation process and pattern

By studying the structural evolution of Moxi-Gaoshiti area and comparing the oil sources, with reference to the accumulation phases and evolutionary history of source rocks, it is suggested that the gas reservoir of Longwangmiao Formation in Moxi-Gaoshiti area is primary oil-gas pool, with the accumulation model of lower-generation-and-upper-storage (principal) and lateral-generation-and-storage (supplementary). The reservoir is characterized by early accumulation, multi-phase fillings, late cracking and later adjustment. Fig. 9 shows the evolutionary history of the accumulation concretely. Controlled by the Leshan-Longnvsi palaeohigh, the Longwangmiao Formation experienced a regional shoaling while depositing, and formed early quality shoal-facies reservoir [2] (Fig. 9a). Hydrocarbon began to fill the porous and permeable reservoir of Longwangmiao Formation as Qiongzhusi source rocks entered oil generation window in the end of Silurian, and formed the first phase of paleo-oil reservoir (Fig. 9b). However, the paleo-oil reservoir was small in scale. Afterwards, with the structural uplift of the Caledonian movement, Qiongzhusi source rocks eventually stopped generating hydrocarbon. During the Caledonian-Hercynian period before the Permian deposition, the Longwangmiao Formation-Silurian strata were subject to denudation in the study area, and as a result, the first phase of paleooil reservoir was damaged (Fig. 9c) and the original hydrocarbon eventually became carbonaceous bitumen after being oxidized in



Fig. 5. Distribution characteristics of paraffin hydrocarbon (limestone data of Permian Longtan Formation quoted from J. Li et al., 2013 [19]).



Fig. 6. Characteristics of salt-water inclusions of Longwangmiao Formation, Moxi-Gaoshiti area. (a) Well Moxi 12, 4632.61 m, plane polarized light, (b), (c) Well Moxi 13, 4615.79 m, (b) plane polarized light, (c) fluorescent light.



Fig. 7. Homogenization temperature of inclusions of Longwangmiao Formation, Moxi-Gaoshiti area.

the supergene zone [20]. In the mean time, the early shoal-facies porous and permeable reservoir of Longwangmiao Formation suffered the transformation by weathering karstification, and formed large amounts of dissolved pores and signs of early carbonaceous bitumen cut by vadose silt were found in the pores (Fig. 10). In the Permian period, the earth's crust sank again. After taking the deposition of the overlying strata, Qiongzhusi source rocks began to generate hydrocarbon again and reached its peak in late Permian. As a result, the second-phase of palaeo-oil reservoir was formed (Fig. 9d). The hydrocarbon generation this time lasted longer, contributed more to the filling and gave rise to a larger scale paleo-oil reservoir than the first phase. In the late Triassic-Jurassic period, the lower Permian source rocks began to mature, and hydrocarbon produced by this filled into the Longwangmiao reservoir laterally through the weathered crust unconformable surface between Permian and Longwangmiao strata, further enriching the second-phase of paleo-oil reservoir. During the Cretaceous period, the Longwangmiao liquid hydrocarbon began to crack into gas, which in-situ



Fig. 8. Depositional, burial and thermal evolution history map of Well Gaoshi 1.



Fig. 9. Gas accumulation pattern of the Longwangmiao Formation, Moxi-Gaoshiti area.



Fig. 10. Carbonaceous bitumen cut by vadose silt (Well Moxi 12, 4620.0 m).

accumulated into gas reservoir (Fig. 9e). The liquid hydrocarbon from Qiongzhusi source rocks left behind carbonaceous bitumen while the liquid hydrocarbon from Permian source rocks left behind oily bitumen, which eventually form the phenomenon of the two kinds of bitumen co-existence.

6. Conclusion

(1) Two kinds of bitumen were found in the Longwangmiao Formation in Gaoshiti-Moxi areas, namely non-fluorescing carbonaceous bitumen and fluorescing oily bitumen. By comparing them with the biological marker compound and the distribution characteristics of n-alkanes of Qiongzhusi source rocks and Permian source rocks, the primary source rocks of Longwangmiao Formation were from Qiongzhusi Formation while the contribution from Permian source rocks was insignificant.

(2) By analyzing inclusions, the Longwangmiao oil & gas reservoir in Gaoshiti-Moxi areas is characterized by three-phase fillings, namely Caledonian-Hercynian filling, Per-mian—Triassic filling and Triassic-Jurassic filling. Caledonian-Hercynian filling was small in scale and produced the first-phase of paleo-oil reservoir that soon destroyed by Caledonian movement uplift; the large-scale Permian filling gave rise to the second-phase paleo-oil reservoir and the Triassic-Jurassic filling, where Permian source rocks filled

complementarily, enriched the second phase of paleo-oil reservoir. During the Cretaceous period, the paleo-oil reservoir cracked into gas reservoir and eventually left behind carbonaceous bitumen and oily bitumen.

Acknowledgements

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