Introduction

Eogenetic interlayer karst and Epigenetic buried hill karst are developed in the carbonate strata of the middle Permian Maokou Formation in southern Sichuan, which is affected by the uplift of Emei mantle plume or the sea level decline and uplift exposure during the global ice age. In the meantime, the presence of multi-episode tectonics movement, the pore fluid properties and diagenetic environment are complex and changeable. Previous studies were short of analyze the control effect of multi-stage diagenetic fluid on reservoir from the perspective of micro-geochemistry, and the coupling relationship between diagenetic fluid and hydrocarbon fluid.

This paper is based on the core and rock section of middle Permian Maokou formation in southern Sichuan, cathodoluminescence, carbon and oxygen isotopes, rare earth elements analysis, fluid inclusion analysis and Raman method were used. The characteristics of macrocosmic petrology and microcosmic petrology and geochemistry are comprehensively studied, and then the multi-stage evolution process of diagenetic fluids and the diagenetic environment of Maokou Formation at each diagenetic stage is discussed.

Geological Setting

The southern Sichuan region is located in the southern sichuan basin, it includes: southern of center-uplift high steep structural belt of southeastern Sichuan, center-uplift low steep dome zone of southern Sichuan and paleo-uplift gentle structure zone. Maokou Formation (corresponds to the Guadalupian) parallel unconformable contact with the overlying Upper Permian Longtan Formation, conformable contact with Middle Permian Qixia Formation. The Maokou Formation is divided into four members from bottom to top, the 1st and 2nd Member of Maokou Formation are divided into three sub-member from top to bottom, most of the area is denuded the top strata of Maokou Formation.

Diagenesis

Grainstone includes: high energy grainstone and low energy grainstone. Calcite cement includes: pectinate calcite, isometric texture calcite, coarse and microcrystalline calcite. The dolomite in southern Sichuan have two types: speckled in limestone matrix, and fill along structural fractures or solution fissures.

Figure 1 Characteristics of reservoir and diagenetic in Maokou Formation of southern Sichuan
A.B20, P₂m₃B, 3655.26m, sparry bioclastic limestone; B. B35, P₂m₃B, 3579.08m, micrite bioclastic limestone; C. Q13, P₃m, 3556.644m, micrite bioclastic limestone; D. B30, P₂m₃, 3142.2m, bio-moldic pore, pectinate calcite and isometric texture calcite; E. B20, P₂m₃B, 3625.37m, calcite filled fracture; F. B20, P₂m₃B, 3661.16m, Orange-dark brown cathodoluminescence belt calcite; G. B20, P₂m₃B, 3658.90m, euhedral dolomite(D) and 3rd stage calcite filled cave; H. B20, P₂m₃B, 3649.3m, dissolution pores, serrated dissolution subhedral micritic crystals and asphalt.

Contemporaneous seawater cementation precipitates unstable pectinate high-Mg or aragonite cementation (CC1) on the edge of biological particles in Maokou Formation. Formation-atmospheric hybrid water dissolves biological cavities such as foraminifera, micrite calcite fills the pores in the
wall of biological cavity to forming micrite envelope, and seepage powder sand is filled in the cavity with the increase of buried depth. In the Eogenetic stage, fresh water isometric calcite (CC2) filled the cavity of biological body to form the geopetal structure. Epigenetic exposure occurred in the southern sichuan area at the end of the middle Permian, the early unstable calcite cement was dissolved and formed into layer-regolith karst and tensile fractures. The stratum settled into the middle - late diagenetic stage again, stylolite which caused by pressure solution cut isometric texture calcite (CC2), bioclasts are broken and arranged in a directional manner under intense compaction. At the same time, coarse sparry calcite (CC3) and idiomorphic and hypidiomorphic (E-D and SE-D) dolomite are filled in karst caves and fractures, CC3 was cut by microcracks which filled with microcrystalline calcite (CC4) (Liu Hong, 2016) (Figure 1).

Rare Earth Element and Carbon and Oxygen Isotope

a. Rare earth element
Limestone of Maokou Formation of southern Sichuan: $\Sigma$REE+Y is 1.72×10^-6-8.33×10^-6, $\Sigma$LREE/$\Sigma$HREE is 0.33-0.90, REE pattern is left-oblique. Y/Ho is 35.54-80.05, REE pattern is similar to the characteristics of seawater at that time. but part of simples’ Y/Ho is less than 44, it shows that the limestone is slightly modified by non-marine source fluids of Maokou Formation (Planavsky N)(Figure 2).

Figure.2 REE+Y distribution type of limestone of Maokou formation in southern Sichuana. A. marine sources limestone; B. limestone modified by hydrothermal fluid.

b. Carbon and oxygen isotope
(1) Grain limestone
Limestone of Maokou Formation can be divided into two types based on the characteristic of $\delta^{13}$C: $\delta^{13}$C is 3.48-4.63‰, $\delta^{18}$O is -8.26--10.72‰ of type A, $\delta^{13}$C is -1.03-2.56‰, $\delta^{18}$O is -6.89--9.71‰ of type B. Characteristic of $\delta^{13}$C of type A and seawater at the same period are basically the same, it shows that the carbon source is mainly inorganic carbon reservoir (marine carbonate) (Alexander B W, 2008). Characteristic of $\delta^{13}$C of type B is obviously negative compared with the seawater of the same period, it shows that the sedimentary and diagenetic processes were modified by non-marine source fluids.

(2) Cements
Fracture infilling calcite cements of Maokou Formation: $\delta^{13}$C is 2.42-5.46‰, $\delta^{18}$O is -11.83--8.18‰, $\delta^{18}$O have a significant negative excursion. Solution fissure infilling calcite cements: $\delta^{13}$C is -1.14-5.85‰, $\delta^{18}$O is -11.25--7.49‰.

(3) Fluid inclusion
The distribution of homogenization temperature of calcite cements coeval brine inclusion of Maokou Formation are studied. CC2: Th1≤85°C; CC3: 85°C<Th3≤170°C; CC4: Th4≥170°C. The homogenization temperature of dolomite coeval brine inclusion is Th4≥170°C which produced at the same time as the middle diagenetic stage CC3. Primary oil inclusions were found in CC3, 76.9°C<Th≤91.6°C; the homogenization temperature of its coeval brine inclusion is
79.1°C<Th≤99.5°C; the homogenization temperature of 4th period coeval brine inclusion is 95°C<Th≤187°C, there are multi-stage gas reservoir forming in middle-late diagenetic of Maokou Formation.

Discussion

a. Seawater, fresh water and deep hydrothermal fluid influence the Diagenetic evolution

Based on the $\sum$LREE/ $\sum$HREE, $\delta$Ce, $\delta$Pr, $\delta$Eu and Y/Ho of rare earth element, REE pattern was divided to two types. One is the sea source limestone: $\sum$LREE/ $\sum$HREE<1, loss of light REE, 76.9°C<$\delta$Ce≤91.6°C, 76.9°C<$\delta$EuCe≤91.6°C, all showed negative anomaly. Another is hydrothermal modified limestone, $\sum$LREE/ $\sum$HREE<1, light REE differentiation reduced, Y/Ho of most of simples less than 44, and $\delta$Eu have light negative anomaly or significant positive anomaly.

The study of carbon and oxygen isotopes demonstrate that hydrocarbon fluids and atmospheric water influence diagenetic evolution, the decrease of sea level and the action of microorganism can lead to the increase of organic carbon oxidation degree in the fluid and $\delta^{13}$C of marine carbonate have a negative anomaly. Tensile fracture which formed in Dongwu Movement and deep large fault which formed in Indosinan-Yanshanian both provide space for atmospheric water eluviation.

b. Prosses of multi-stage paleofluid hydrocarbon charging evolution

In the eogenetic period of Maokou Formation, dissolution moldic pores were formed in the biological particles under the atmospheric water and primary seawater mixing. At the same time, tensile fracture and dissolved vugs were formed at upper strata of Maokou Formation by Dongwu Movement. Maokou Formation was rapidly subsided into the middle diagenetic again in late Permian - early Triassic, high-amplitude sutures and clay laminae caused by pressolution have destructive effects on the reservoir. In the middle diagenetic stage, the paleogeothermal temperature of Maokou formation exceeded 85°C. Silurian source rocks got maturely in Early Triassic, and crude oil charged upward to Maokou formation to form paleo-oil reservoir. Paleo heat flow value was significantly increased to 70-100 Mw/m² in late Permian - early Triassic, it created a condition for the precipitation of dolomite in the middle-buried stage. But the thermal events that satisfy the dolomitization have shorter duration((257±3)Ma-(263±5)Ma), dedolomitization occurred when the geothermal temperature decreased rapidly to improve reservoir quality.

Figure.3 Paleofluid evolution of middle Permian Maokou formation of southern Sichuan
Middle-late Triassic (248 Ma - 221.5 Ma), 1st gas charging occurred in Maokou Formation. The paleotemperature of Maokou Formation in the early middle diagenetic stage did not reach the conditions of crude oil pyrolysis (Th of calcite brine inclusion is between 98.5 °C - 129.6 °C), 1st gas charging stage was the pyrolysis gas from lower strata migrated to Maokou formation along the fault. Since the Jurassic, the intensive tectonic compression by Yanshan Movement and hydrocarbon charging caused the expansion fractures in the reservoir. It captured the charging pyrolysis gas of Maokou Formation which from paleo reservoir of early middle Jurassic (208 Ma - 163 Ma) and end of the early Cretaceous (128 Ma - 100 Ma) in late diagenetic stage. Tectonic uplift occurred in the southern Sichuan during the Himalayan period, and fractures formed in the early calcite cements. Deeply acidic formation water precipitated in the fracture and 4th gas charging associated with it, and the gas was the migration pyrolysis gas which from early paleo reservoir accumulated (Figure 3).

Conclusion

(1) Maokou Formation, southern Sichuan Basin, was occurred “penecontemporaneous seawater cementation → eogenetic mixed water cementation → epigenetic atmospheric freshwater dissolution → phyllomorphic formation water cementation, metasomatism and acidic-fluid dissolution”. Origins of sedimentary and diagenetic fluid include seawater, atmospheric freshwater, hydrocarbon fluids and thermal fluids.

(2) The anomalous thermal of the Emei mantle plume caused multiple effects on the evolution of karst reservoirs and fluids of Maokou formation: during the Middle Permian, mantle plume caused massive erosion of the Maokou formation, and Maokou formation formed weathering crust solution fissures by weathering leaching; during early Triassic, the anomalous thermal of the Emei basalt eruption during early Triassic, it caused the Silurian source rocks maturely, crude oil injected upward along the fault to the Maokou formation, and it also caused the brief dolomitization of Maokou formation. Organic acid dissolution and dolomitization inhibits the filling of formation water cement which are play constructive roles in Maokou Formation reservoir.

(3) Maokou Formation was experienced multi-phase oil and gas charge: gas from middle - late Triassic lower strata charged to the dominant karst reservoir of Maokou formation; a large amount of gas charging occurred during the pyrolysis of paleo reservoirs in early Jurassic-early Cretaceous Maokou formation, deep buried environment coarse-grained calcite cement and residual asphalt blocks reservoir space; until the Himalayan period, tectonic faulting improves the reservoir space again, early gas adjustment which migrated to karst reservoirs associated with fractures charged the reservoirs again.

Reference

