LI ZHIWEN
and
GUO JIANHUA

Prospect of Middle Ordovician Yanxi formation black shale in South Hunan province

Black shale is widely distributed in Central Hunan. However, due to low exploration level and lack of petroleum geological data, it lacks complete evaluation.Based on the comprehensive investigation and analysis of the geological characteristics of Yanxi formation (O_2) and the sedimentary evolution of Ordovician in this area. it is found that the black shale of O_2 is characterized by great thickness (from 40 m to 135 m) and high level of organic carbon (from 0.87% to 6.20 %). Based on illite crystallinity (IC) analysis, the thermal maturity of O_2 is at the stage of mature to over-mature (1.83%<Ro<3.17%, and the average value is 3.15%), and the epizone (0.12<IC<0.252, mostly 0.12 ~ 0.192) is not beneficial for producing gases. However, the O_2 is still a gas-bearing stratum with great potential of shale gas, and the difficulty of the shale gas exploration in this area lies in the elusive and complicated structure.

Keywords: *Middle Ordovician, Yanxi formation, shale gas, sedimentary evolution.*

Introduction

Shale is a type of fine-grained sedimentary rock, which forms from the compaction of silt and clay-size mineral particles while shale gas is a type of natural gas that is trapped within shale formation [1]. Shale gas has become an increasingly important natural resources in the United States since the beginning of this century, and great attention has been paid in other parts of the word. The production of shale gas accounted for 1% of natural gas in 2000 in the United States, and reached 28.23% by 2016. It is estimated that the world's largest shale gas accumulation area is in China, most of which lies in three basins, namely Sichuan basin, Tarim basin and Yangtze Platform, which accounts for 89% of the estimated national reserves. The shale gas exploration in China was initiated in 2009 with the shale gas recoverable reservoirs being about 31 trillion m^3 (according to the Energy Information Administration); however, little progress was made in the first three years. China produced 200 million m^3 (7.1 billion cubic feet) of natural gas from shale formation in 2013, which was less than 0.2% of China's total natural gas production. With breakthroughs in Fuling, Chongqing, Changning and Weiyuan in Sichuan in 2014, China has produced 1.5 billion cubic meters of shale gas.

The whole industry has recovered. Under the guidance of successful experience and previous work, the researchers have widely targeted the margin of Yangtze platform, going deep into Paleozoic and even Proterozoic.

As the arrival of the success in Yichang, Hubei Province, a new window of the shale gas was opened in southern China.

1. Geological setting

Hunan province is located on the southeast edge of Yangtze Platform in southern China; and the research area received sediments from different units including platform, platform edge and basin from Neoprotezoic to Early Paleozoic. Northwest area of Hunan province belongs to platform sedimentary unit, while the southern Hunan belongs to platform edge and basin unit, which mark the frontier of a super-convergent regime. Within the super-convergence domain, the compressional structure in the central south of China block is mainly characterized by shortening, thrusting and decollement. Different types of sediments together with the complex tectonics constitute the various geological conditions and laws in Hunan province. As the precursors have concluded, there develop 5 main source rocks and 3 secondary rocks, which are mostly black shales in the research area: Lower Cambrian Niutitang Formation ($□_1n$), Lower Silurian Longmaxi Formation (S_1l), Devonian, Lower Carboniferous, Upper Permian and Sinian, Upper Ordovician Wufeng Formation (O_3w), Lower Triassic. Taking the caledonian unconformity as the boundary, the source rocks in these area are divided into the upper group and lower group. The source rocks of the platform area are comprised rocks of both upper and lower groups; whereas the source rocks from platform edge to basin area only contain rocks of the upper group. Precursors believe that the Pre-Devonian source rocks in southern Hunan province have metamorphosed and cannot generate hydrocarbon anymore;

Messrs. Li Zhiwen and Guo Jianhua, School of Geoscience and Info-Physics, Central South University, Key Laboratory of Metallogenic Prediction of Nonferrous Metals and Geological Environment Monitoring, (Central South University), Ministry of Education, 410083, China. email: 281783800@qq.com

thus take the caledonian unconformity as the deadline of hydrocarbon. Therefore, the former explorations did not reach the formation underlying the Devonian in southern Hunan province; even though thick black shales which are rich in total organic carbon (TOC) develops in the south area of Hunan and part of the Yangtze Platform edge. Niutitang formation covers most areas of the southern China and develops black shale of 150~625m thick with a high TOC content ranging from 0.55% to 19.9%. Its equivalent vitrinite reflectance value (Ro) that is converted from bitumen reflectance [2] in early Paleozoic when there is no vitrinite at all, is generally high, and usually exceeds 3.0% and reaches 4.0% in some cases [3-4]. It has been proved in Fuling (the first industrialized shale gas field in China) that the shale whose Ro value reaches 3.5% can still generate gas [5-6].

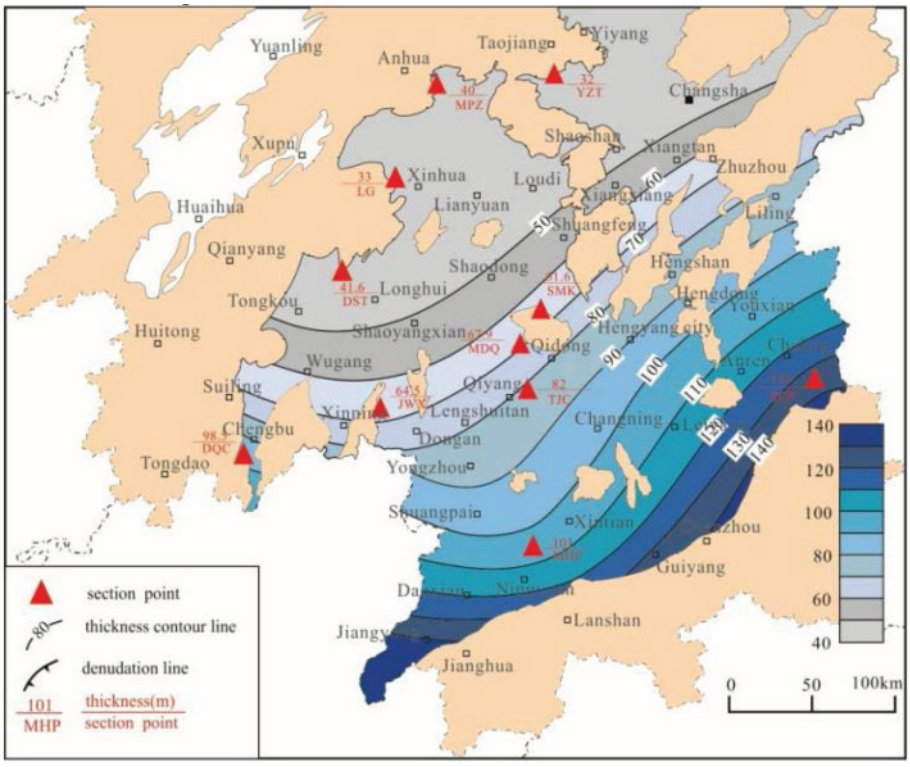

Fig.2 Shale thinkness isoline of middle Ordovician Yanxi formation in Central Hunan

As the platform edge has experienced multistage tectonization, fewer people believe that the lower Paleozoic still has oil-bearing potential after suffering the regional metamorphism [7-8]. However, the metamorphism in southern Hunan belongs to low-temperature dynamic metamorphism, and has anisotropic properties. The precursor once said, "in Hunan province, the upper layer is too unpredictable and the lower layer is too old." This experience is applicable to conventional exploration; but we have to turn to new guidelines for non-traditional exploration. So far, most shale gas in China has been found in the black shale in Longmaxi formation of Yangtze Platform. The thickness of Longmaxi formation black shale in the platform unit of the research area (northeast Hunan) is less than 20m; For shale gas development, the thickness of high-quality shale should not be less than 30m. In other words, the Lower Paleozoic shale gas in Hunan is located in deeper strata, not Silurian.

2 Materials and methods

2.1 Field work and facies description

Except for the Liutitang formation [$\square_1$n], another shale dominant stratum Yanxi formation of the Middle Ordovician (O_2y) is also extensively developed in the central Hunan province. Researchers describe over 18 sedimetological sections (2,000m of cumulative thickness) for facies analysis and collection of biostratigraphic information.

Thickness is the main indicator of depocenter and its resource potential. And the shale in the southern area is much thicker than that in the north. The thickness of the northern part, such as Xinhua, Yiyang etc. is less than 40m, while the thickness of the southern area around Lingling, Qidong and Hengyang ranges from 65

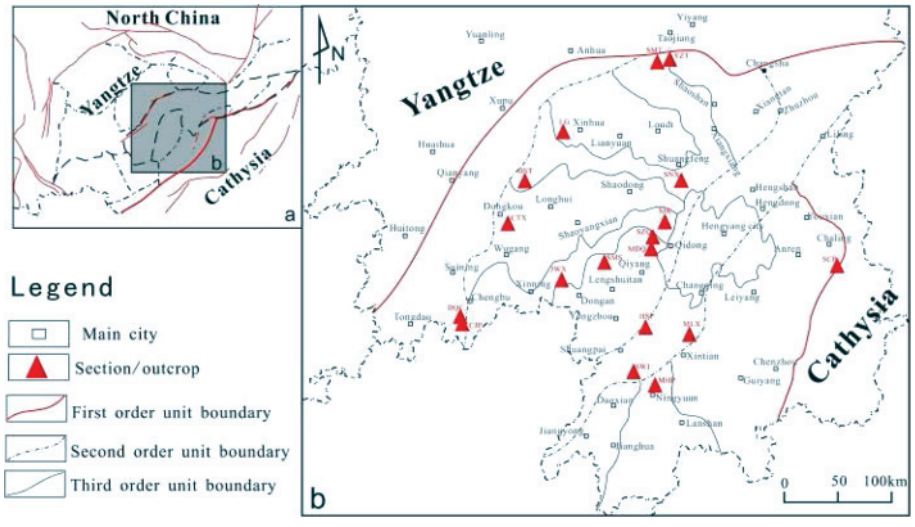

Fig.1 Regional location of central Hunan area and main outcrops of Yanxi formation

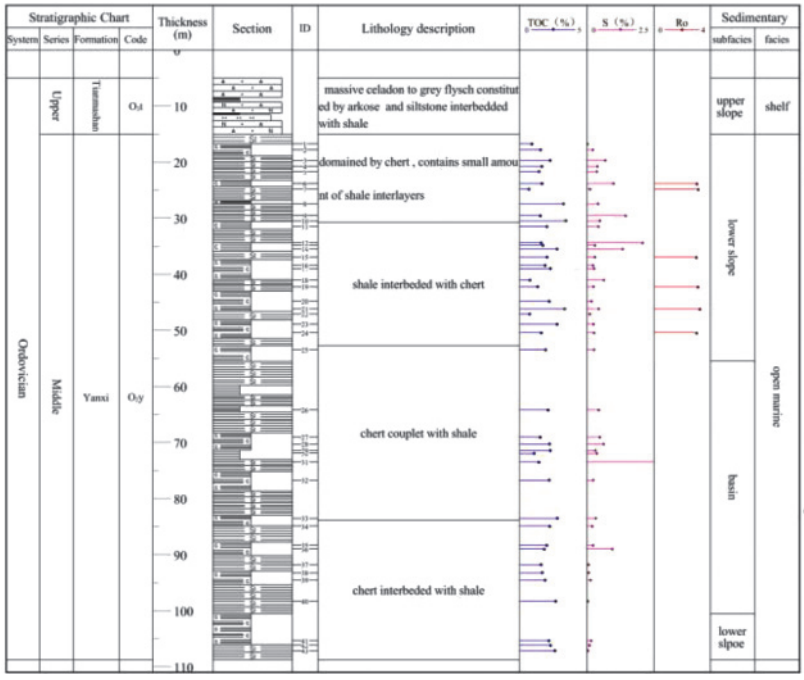

Fig.3 Comprehensive stratigraphic column of O_2y in MHP

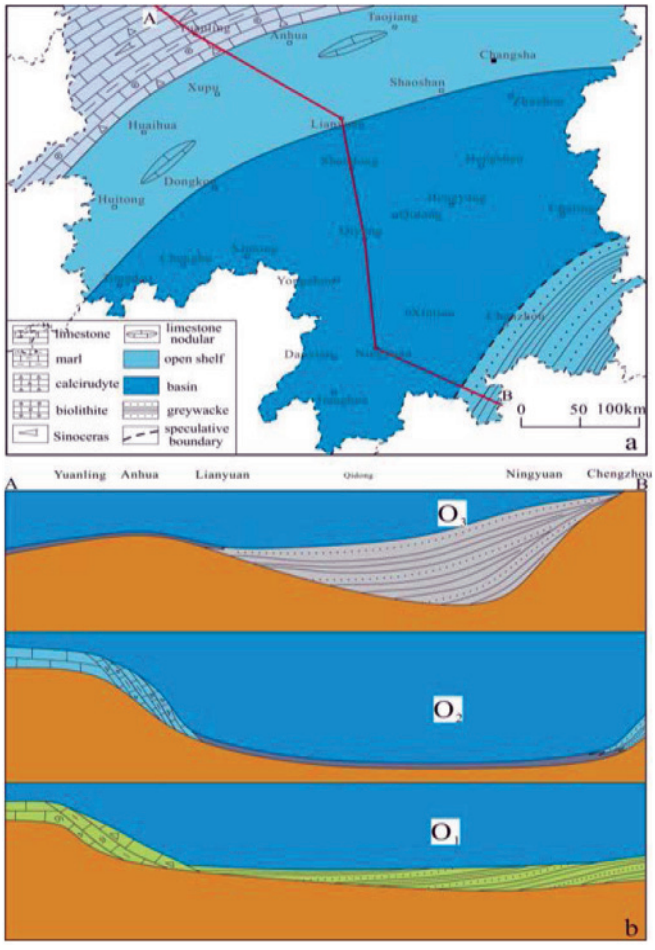

Fig.4 The sedimentary facies plan of O_2 (a) and the evolution model of Ordovician(b) of central Hunan

to 95m. The thickness in the Chengbu, Ningyuan, Chaling, etc. exceeds 80 m, and it even reaches 135 m in Shichangpin of Chaling County (Fig.4). The effective thickness refers to the total thickness of the high quality shale in a continuous shale section. Only if the effective thickness of the shale is big enough (>30m), an industrial gas reservoir can form in the shale.

The facies which are grouped under two main depositional environments (shelf and open marine) constitute different sets of facies associations; however, the difference between them is subtle. The sedimentary facies in the Middle Ordovician study area are mainly open marine facies, which changed to platform margin in the north and to clastic cliff shelf in the south (boundary is unclear).An extensive transgression hits south China during the Middle Ordovician, and the open marine facies can be divided into lower slope and basin subfacies based on their different rock components. The rock assemblage of the two subfaceis is composed of carbonaceous shale and chert, the main difference is that the content of chert is higher in the basin.

According to the rock association and facies evolution in different strata of the Ordovician, it is found in section AB that: (1) The basin which is affected by the terrestrial sources is shallow and is mainly formed by deposited fine clastic rocks, such as silty shale and siltstone in early Ordovician; while the carbonate platform is developed in the north. (2) The water deepens in the middle Ordovician and shows an obvious retrograding feature, the areas to the north of Anhua and Xupu still develops carbonate platform, while deep water shelf develops to the south of the belt of Dongkou, Lianyuan and Shanshan; and the sediments are composed by the cherts coupled with shales and partly developed limestone lens. The extensive area from Lianyuan to Chenzhou develops the basin facies, and sediments are mainly composed of deposited cherts interbeded with shales of different thickness. The region that is to southeast of Chenzhou is speculated to be clastic shelf, where the sediments are composed of fine detrital deposit, and which are similar to that of early Ordovician. (3) The late Ordovician is the reversal stage of the central Hunan basin, and this area enters into a foreland basin evolution episode along with the squeezing of the blocks. The foredeep develops and gradually moves northward, and finally forms a regional mélange with northward thrusting and southward thickened fault blocks. And the original basin area is filled up with medium to coarse clastic sediments, the deep shelf region bulges up, while the

area to the north of Yuanling with carbonate deposit sinks and deposits into black siliceous and carbonaceous shale, which is totally different from that in the southern part (Fig.4).

2.2 Geological analysis

Total organic carbon (TOC) is used for characterizing the abundance of organic matters [7]. It has a positive correlation with the gas-bearing property, and determines the amount of organic pores and the adsorption capacity of the shale, which is another key index to indicate whether shale gas can accumulate within a reservoir. The TOC of 139 samples in O_2y is assayed for evaluating the potential of the O_2y (Table 1). It is found that the TOC of the black shale in O_2y with an average value of 2.12% is generally high, with the maximum value being 8.17%; and the heterogeneity of the TOC is mainly caused by the sedimentary facies distribution and eustacy, indicating that the O_2y belongs to the high-quality source rock. However, its TOC values are different from the north to south. The favourable areas with the highest values of TOC are located in Longhui, Dongkou, Qidong and their vicinity; and another favourable area is along the line of Lingling, Ningyuan and Chengbu, where the TOC value ranges from 1.97 to 2.94%. Above all, the black shale in O_2y of the research area which is rich in organic matters and has a high TOC value is good source rock for the shale gas; and the TOC value is high in the contact zone of shale and chert as well.

The thermal evolution of the organic matters is a decisive factor for the generation ratio of hydrocarbon; while it is also in a close relation with the burial history, regional structural and thermal events. The residual thickness of the Paleozoic reaches 5,000~10,000 m, and the structural activities and thermal events are very active in the Late Paleozoic. The burial and preservation conditions, as well as the thermal evolution degree of the O_2y in this area are unpromising for shale gas judging from the appearance, since the graphite deposit occurs both in the Lower Devonian of Huangcai County of Ningxing in the north and the Heye County of Guiyang in the south of the research area, and the metallogenic temperature is above 1,200°C.

The research area has undergone the Caledonian, Hercynian, Indosinian and later movements, and re-deformed thereby. There are various structural sinks and uplifts existing in different tectonic divisions; and the heterogeneity is obvious since the burial depth is not very deep and the burial time is not very long in many areas. Based on the test of bitumen reflectance (Rob) of 45 outcrops, the Rob of the samples of O_2y ranges from 2.27% to 5.46%, with the average value being 4.62%. According to the correlation between bitumen reflectance and vitrinite reflectance (Ro) (Jacob, 1985), the Ro value of the O_2y in this area is from 1.83% to 3.77%, with the average value being 3.15%, which is much lower than that from former cognition. The Ro of the shale in the shale gas fields of the United States generally

Table 1: Statistics of TOC and Ro of outcrops of O_2y in central Hunan

Section	TOC (wt.%)	Ro (%)
CJP	2.32 / 0.75 - 4.05 (20)	3.13 / 3.06 - 3.22 (4)
DJC	1.38 / 0.55 - 2.77 (20)	3.10 / 3.04 - 3.14 (4)
CTX	4.42 / 1.16 - 8.17 (3)	2.27 / 1.83 - 3.30 (3)
SKC	1.58 / 1.58 - 1.58 (1)	3.16 / 3.16 - 3.16 (1)
DST	6.20 / 3.79 - 7.57 (5)	3.71 / 3.66 - 3.77 (5)
SNX	1.96 / 1.85 - 2.13 (3)	6.20 / 3.79 - 7.57 (5)
MDQ	4.46 / 3.28 - 5.79 (7)	3.62 / 3.39 - 3.84 (6)
SJK	3.14 / 3.02 - 3.25 (2)	2.93 / 2.75 - 3.10 (2)
SMS	1.97 / 1.96 - 1.97 (2)	2.93 / 2.75 - 3.10 (2)
JWX	1.15 / 0.15 - 2.14 (31)	3.06 / 3.00 - 3.15 (5)
WSP	2.94 / 2.90 - 2.97 (2)	3.21 / 3.14 - 3.28 (2)
MHP	1.96 / 0.69 - 3.44 (43)	3,.15 / 3.02 - 3.27 (6)

Symbol: average (value) / minimun – maximum (value) (number)

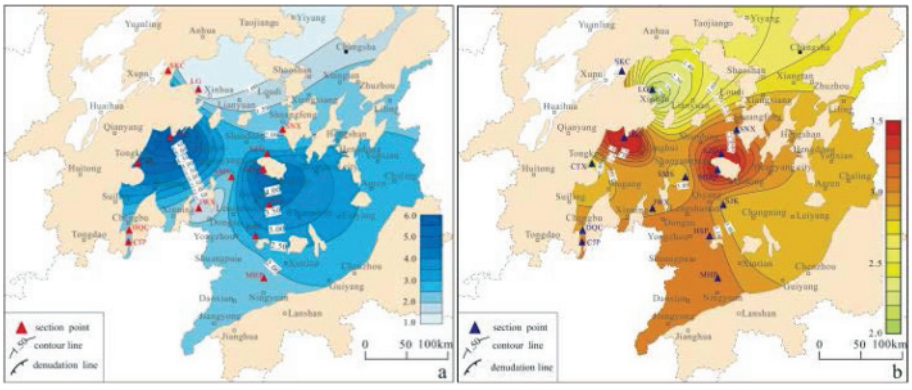

Fig.5 Organic carbon content (a) and vitrinite reflectance (b) of O_2y black shale in central Hunan

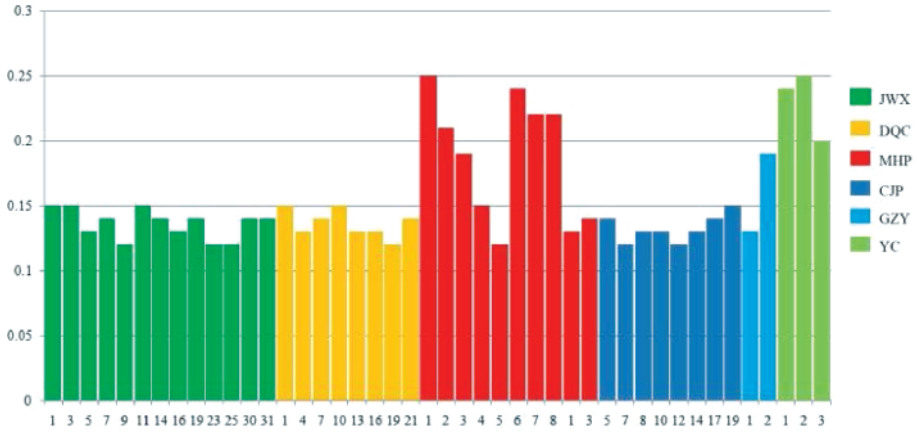

Fig.6 IC of the outcrops in central Hunan and Yichang (YC) area

exceeds 1.3%; in particular, it reaches 4.0% in southern West Virgina of the Appalachian basin. Moreover, the gas only produces in the over-matured areas; high thermal evolution degree (Ro>3.0%) is not the principal factor that restricts the accumulation of the shale gas (Montgomery et al., 2005); and the Ro of this area is below the ceiling if a certain distance is maintained from the sags and intrusions. Xinning, Qidong and other areas are relatively better for exploration.

The thickness, TOC, maturity, and kerogen type are common geochemical indexes for evaluating the potential of shale; but they seem specious with regard to old formations (especially prior to Silurian). Van Krevelen chert is widely used for classifying the kerogen type, but it cannot work well for the post-mature source rocks [8]. Vitrinite can record the highest temperature that a stratum has ever suffered, which can reflect the maturity of the source rocks; but the constituent and optical performance under the high temperature of the bitumen vary, and the convert formula between Ro and Rob is not persuasive. Illite crystallinity (IC) [9] is widely applied in "very low grade metamorphism" (VLGM) and in petroleum exploration so as to detect diagenetic stages. The VLGM zone can be divided into three grades based on Kubler index: diagenetic zone ($IC>0.42_2$), anchizone ($0.25< IC<0.42_2$) and epizone ($IC<0.25_2$); among them, the anchizone corresponds to hydrocarbon generation phase.

The testing results of the IC show that the Yanxi formation has entered into the epizone and O_2 has lost its potential, which is consistent with the precursors' concludions. But according to the research report of the copper ore in Ningyuan area, the ignitable gas flow is found in the Cambrian during ore drills in the Ganziyuan (GZY) area. The Middle Cambrian which consists of silty slab and carbonaceous slab is overlain by the sandstone of Lower Devonian. Its thermal maturity is much higher, but its IC is lower than 0.19_2. The qualified aeration zone in central Hunan is contained in the black shale of the Upper Paleozoic, including Devonian, Carboniferous and Permian. These strata are eroded away after Indosinian in GZY area, where the faults intersect with each other. The natural gas found in the ore drills is shallower than 200 m, so there is little chance that the gas comes from the Upper Paleozoic, but is more likely to come from the Cambrian shale or deeper strata. Comparing with the Yichang area where shale gas is found in Lower Cambrian ($0.20< IC< 0.25_2$), the IC in central Hunan ($0.12< IC< 0.25_2$, mostly 0.12~0.19) is lower than that of Yichang area, and only the samples from MHP profile are identical to that Yichang area. It proves that the shale in epizone cannot generate gas but can restore some gas. Furthermore, anisotropism occurs in Yanxi formation, and the IC of two testing samples in MHP is in anchizone. To sum up, the black shale in O_2y is a prospective stratum.

2.3 Analysis of preservation condition

The exploration achievements of the Sichuan basin and surrounding areas reveal that the preservation condition is the key factor of the shale exploration in China. The favourable structural styles include box faulted anticline, gentle plate-like syncline, symmetric broad anticline and latericumbent syncline. The structural remodelling extent of the central area of Hunan is higher than that of areas, like west Xuefeng propagation thrust and Qiyue-Huaying high-steep folded belt, the continuity of the stratum is worse, while the interlayer slide and large scale thrust are very developed in this area. Thus, the prerequisite for searching a relatively stable district in the central area is to understand the prototype basin and the tectonic transformation process in the early Paleozoic.

Through analysis of the lithological association and sedimentary facies, it is identified that the central area of Hunan is a deep water basin formed in the middle Ordovician, the region to the north develops shallow carbonate platform, while deep water shelf develops to the south. The central area of Hunan basin is almost filled up by the turbidite in the late Ordovician, which is not coordinated with the normal sedimentary sequence and facies association. Moreover, nearly 100 km of strata are reversed in areas from Daoxian to Shuanpai, which cannot form in ordinary reversed anticlines. Besides, the O_2y contains abundant radiolarians, which indicates the pelagic deposits. Combining the pelagic sediments, turbidite and reversed strata, the authors believe that the Lower Paleozoic, especially the O_3t of the Upper Ordovician, are not normal sedimentary strata, but a set of mélange that formed in the subduction process between Yangtze and Cathysia blocks. The massive reversed strata, repetition and sedimentary facies skipping corroborate the idea of mélange. Thus, the difference in thickness of the

Ordovician from the north to south, especially for the O_2y and O_3t, may be caused by the tectonics.

3. Discussion

Big progress has been made in shale gas exploration in south China. Industrial discovery has been achieved in periphery of Sichuan basin, like Chongqing, Guizhou and Hubei, and the exploration blocks are gradually expanded from the basin to the orogen [12-14]. In spite of abundant hydrocarbon as identified in the drills, the industrial breakthrough is still pending for searching from somewhere in Hunan province. As a new stratum with high potential of shale gas, the O_2y is the key factor for shale gas exploration in Hunan province.

Water of a certain deep can supply a reducing environment, which is good for the preservation of organic matters [15-16]. The main discovery layer named Longmaxi formation of the Lower Silurian (S_1l) deposits in the deep shelf facies; and the O_2y develops in deep basin, where the water is too deep to reserve organic matters, a lot of them are dissolved in the water column, and the chert layer accounts for a large proportion of whole formation. The TOC of the layer is lower than that in the S_1l, but it still belongs to good source rocks. The limiting factor of the O_2y is that its sedimentary water is too deep, and it consists of chert coupled with shale in terms of lithology. Even though it is thick enough, the continuity of thickness of the shale is limited. This lithology structure is good for hydrocarbon expulsion, which doubts the remnant gas content in the shale. In addition, though several researchers have proved that the chert of the O_2y in the central Hunan is originated from sediments of rare earth elements [10-11], the limited and simple site cannot represent the whole area. On account of tectonic and thermal activities of multiple stages, a lot of veins are spotted in the outcrops and cores, and alteration is also found in the chert with microscope; another genesis is that the hydrothermal fluids invade through decollement layer within the O_2y and siliconize the shale. However, if the chert is originated from hydrothem, the exploration potential of the O_2y will decrease.

It is proposed that the Lower Paleozoic in the central area of Hunan belongs to mélange, which further discounts the potential of the O_2y. While searching for gas resources in the mélange belt, not only the attitude of the strata and cap condition should be taken into account, but also the attitude and scale of the mélange block. The structure in central Hunan is complicated, the inner structural gradation and style are distinct from other areas; and the underground structural conditions cannot be obtained through ground survey. In other words, it is very hard to effectively evaluate the exploration potential in the absence of geophysical and drilling information in this area.

4. Conclusions

Based on tectoic-sedimentary and geochemical analysis, this paper reaches the following 5 points:

1. The black carbonaceous shale in O_2y in central Hunan is widely distributed with great thickness, and it is a new reservoir of shale gas in Hunan province.
2. The sedimentary facies in the central area of Hunan are mainly comprised deep-water shelf and deep basin, with the region to the north developing into carbonate platform edge deposit, while detrital sediment depositing in the south slope. Squeezed by the Cathysia, the central area of Hunan basin transformed in the Late Ordovician, the type and the filling-up sequence are conversed as well.
3. The O_2y consists of four sub-layers from the bottom to the top, the first layer is composed of chert interbeded with carbonaceous shale, the second layer consists of chert coupled with carbonaceous shale, the third layer, mainly comprised black shale, is the best layer for exploration, and the forth layer is mainly composed of chert .
4. The thermal maturity of $O_2y$is at mature to over mature stage ($1.83\%<Ro<3.17\%$, and the average value is 3.15%), and the IC ($0.12< IC< 0.25_2$, mostly $0.12\sim0.19_2$) indicates that this stratum has entered into epizone stage, which is not beneficial for producing gas. However, ignitable gas is also discovered in GZY area ($0.13< IC< 0.19_2$) and YC area ($0.20< IC< 0.25_2$). Thus, the O_2 is still a gas-bearing stratum with great potential of shale gas.
5. The reversed strata of large scale is related to the plate subduction, the strata in the central area of Hunan subject to this research are mélange rather than normal sediments, which is a great challenge in shale gas exploration.

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ASSESSMENT AND REVIEW OF MAINTENANCE PRACTICES IN THE 4TH INDUSTRIAL REVOLUTION USING THE COGNITIVE ANALYTICS FRAMEWORK

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