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Research article

Electric property evidences of carbonification of organic matters in marine shales and its geologic significance: A case study of the Lower Cambrian Qiongzhusi shale in the southern Sichuan Basin

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Abstract

Searching for some reliable evidences that can verify the carbonification of organic matters in marine shales is a major scientific issue in selecting shale gas fairways in old strata. To this end, based on core, logging and testing data, the electric property of two organic-rich shale layers in the Lower Cambrian Qiongzhusi Fm. and the Lower Silurian Longmaxi Fm. in the southern Sichuan Basin was compared to examine the carbonification signs of organic matters in the Qiongzhusi shale and its influence on gas occurrence in the shales. The following conclusions were reached: (1) the electric property experiment shows that the Qiongzhusi shale in the study area has had carbonification of organic matters. The low resistivity of dry samples from this highly mature organic-rich shale and ultra-low resistivity on downhole logs can be used to directly judge the degree of organic matter carbonification and the quality of source rocks; (2) in the Changning area, the Qiongzhusi shale shows low resistivity of dry samples and low to ultra-low resistivity on logs, indicating that organic matters are seriously carbonized, while in the Weiyuan area, the Qiongzhusi shale shows a basically normal resistivity on log curves, indicating its degree of graphitization between the Longmaxi Fm. and Oiongzhusi Fm. in the Changning area; (3) shale with medium-to-high resistivity is remarkably better than that with ultra-low resistivity in terms of gas generation potential, matrix porosity and gas adsorption capacity; (4) industrial gas flow has been tested in the organic shales with medium-to-high resistivity in the Jianwei-Weiyuan-Tongnan area in the north, where the Qiongzhusi shale is a favorable shale gas exploration target.

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The Lower Cambrian Qiongzhusi Fm. is one of the key marine shale gas exploration and development targets in south China [1-3]. Over the past few years, Chinese geologists have carried out studies on source rock evaluation, shale reservoir characterization and strategic fairway identification for this formation [1-9]. Overall, it is considered that the black shale in Qiongzhusi Fm. has favorable conditions for shale gas

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occurrence, such as wide distribution, great thickness of organic-rich shale (Fig. 1), and high brittleness [1-7]. However, this old shale (about 0.57 billion years B.P) with superhigh thermal evolution degree (R_0 is between 2.5% and 5.0%), has had carbonification of organic matters occurring in the Sichuan Basin and its surrounding areas, making its accumulation conditions and gas bearing properties apparently poorer than those of Lower Silurian Longmaxi Fm. [6-8]. Moreover, the major hydrocarbon generation source materials in Cambrian are low planktonic algaes, without high plant remains [6], and lack of vitrinite and bitumen, leading to

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Fig. 1. Black shale distribution and related well locations in this paper in the lower member of the Lower Cambrian Qiongzhusi Fm. in the southern Sichuan Basin (modified from Ref. [8] in this paper).

bigger differences of $R_{\rm o}$ values of the same rock samples tested in various labs, which usually can't reflect the actual situation of thermal evolution degree, thus it is hard to select shale gas fairway areas for the Qiongzhusi Fm. Searching for some reliable evidences that can characterize the carbonification of organic matters in marine shales and revealing its effect on shale gas bearing properties are major scientific issues in fairway identification in old shale gas plays.

Studies have proved that during the thermal evolution process of source rocks, with the rise of thermal maturity, organic matters are firstly degraded to kerogen; in the changing process followed, the kerogen generates hydrocarbons with continuously increased volatility and hydrogen content, along with decreased molecular weight, and finally forming methane. With the rise of temperature, kerogen constantly changes, so does its chemical compositions, gradually turning into carbonaceous residues with low hydrogen content and finally converting into graphite (namely carbonification) [10]. Currently, at home and abroad, there are few studies on the carbonification during the thermal evolution process of source rocks, and there are no effective geochemical methods to identify the carbonification of organic matters.

A lot more efforts have been put into the study and exploration of the Qiongzhusi Fm. instead of the Longmaxi Fm., in which only four wells have been drilled (numbered A, B, H and I) for shale gas appraisal, mainly distributing in Weiyuan, Changning and Zhenxiong areas in Sichuan Province and Yunnan Province. Other wells (numbered D, E, F, G and J) drilled to the Cambrian shale are all exploration and appraisal wells for conventional oil and gas (Fig. 1). As carbonification of source rocks may lead to low to ultra-low resistivity response characteristics in electric logging curves, based on core, logging and testing data in the Qiongzhusi Fm., we carried out resistivity experiments on dry organic-rich shale samples to find out direct evidences of carbonification of source rocks, and then calibrated the downhole resistivity log data, and characterized the carbonification of organic matter in the Qiongzhusi Fm. in various areas of the southern Sichuan Basin by resistivity responses; meanwhile, by comparing the corresponding properties (such as electric property, lithofacies, geochemistry and adsorptive capacity, etc.) in the Longmaxi Fm., we examined the effect of carbonification of organic matter in different degrees on shale gas occurrence conditions to find out new ways for geologic evaluation of marine shale gas with an ultra-high maturity, and provide geologic foundation for fairway identification of shale gas resources in the Qiongzhusi Fm.

1. The status quo of the study on the carbonification of organic matters in shales

At home and abroad, not much study on carbonification during the thermal evolution process of the source rocks has been reported, and the study methods are limited to microscopic observations, kerogen element analyses and well logging responses etc. [10-14].

Graphite crystals in hexagonal flake structure under microscopic observation are direct evidence of carbonification of organic matters, but it needs microscopes with ultra-high resolution and enough flake-like graphite crystals in rock samples to find out this evidence. The C–C bond of graphite hexagon is only 0.142 nm long, and each net layer is only 0.34 nm apart, but the resolution of a high magnification scanning microscope is generally lower than 10 nm. Thus it is very difficult and infeasible to look for graphite crystals in organic matters with a high magnification microscope.

Kerogen element analysis is an important way to judge carbonification of organic matters. Some researchers (such as Chen Jianping and Cheng Keming [11,12]) thought that the gradual decrease of atomic ratio of H/C with the hydrocarbon generation evolution progress of organic matters was an important and effective index to measure the maturity evolution degree of organic matters. When H/C ratio of marine source rocks declines to below 10% (corresponding to $R_o > 3\%$), the source rocks enter a graphitization stage (i.e., carbonification). This understanding is a deduction obtained by experimental modeling, and the max R_o value in the experimental modeling was 3%. Therefore, it cannot completely reflect the actual maturity status of marine shales in southern China, and also cannot interpret the carbonization degree of organic matters in the Qiongzhusi shale.

Resistivity logging response is an important basis for studying thermal evolution degree of organic matters [13–15], and it is also an indirect evidence for characterizing the carbonization degree of shale [8]. Graphite is a highly conductive mineral, with a resistivity of 8×10^{-6} –13 × $10^{-6} \Omega$ m at ambient temperature. In organic-rich shales of some old formations (such as the Qiongzhusi Fm. and Longmaxi Fm.), the organic matters, between 2% and

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