Advances in Geo-Energy Research

Perspective

Structural evolution and characterization of organic-rich shale from macroscopic to microscopic resolution: The significance of tectonic activity

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Keywords:

Organic-rich shale shale gas structural deformation Sichuan Basin

Cited as:

Gao, J., Li, X., Cheng, G., Luo, H., Zhu, H. Structural evolution and characterization of organic-rich shale from macroscopic to microscopic resolution: The significance of tectonic activity. Advances in Geo-Energy Research, 2023, 10(2): 84-90. https://doi.org/10.46690/ager.2023.11.03

Abstract:

Shale gas exploration and development have taken significant strides in the relatively straightforward intra-basin stability zone and intra-basin weak deformation zone of marine shale in the Sichuan Basin, South China. In addition, the extra-basin strong tectonic modification zones have been actively explored. However, the results have been limited, which reveals the complexity of shale gas formation and preservation conditions in the context of multi-scale geological processes. These tectonic geological conditions have a significant impact on the shale gas content, while it has been difficult to figure out how tectonic deformation modifies reservoir structure and what specific mechanism causes shale gas content anomalies. Based on subjecting geologic samples to combined hightemperature and high-pressure experiments, this study summarizes the tectonic constraint mechanism of shale petrophysical structure evolution and its impact on shale gas storage, reveals the intrinsic connection and mechanism of shale pore-fracture and organic matter, inorganic mineral particle structure evolution and tectonic stress, and identifies the remodeling mechanism of the shale reservoir physical property change. The findings contribute to the theory of shale deformation and gas accumulation, as well as offer a scientific foundation for the exploration of marine shale gas in the complex tectonic zones outside the Sichuan Basin.

1. Introduction

In the past 20 years, organic-rich shale rapidly developed into the most important exploration area for unconventional oil and gas in the world, and shale oil and gas production also realized explosive growth (Zhu et al., 2018). The Lower Paleozoic Qiongzhusi Formation and the Wufeng-Longmaxi Formation are two sets of high-quality hydrocarbon source rocks that are widely developed in the Upper Yangtze area

of south China, and their organic-rich shale sections are essential strata for shale gas exploration and development (Zhu et al., 2019a, 2019b). In recent years, the exploration and research work of marine shale gas focused on the Sichuan Basin and its peripheral areas and made important progress in terms of nanopores (Ma et al., 2015; Guo et al., 2022; Yang et al., 2022), natural fracture network (Zhu et al., 2019a), petrophysical properties (Zhu et al., 2021), gas content (Zheng et al., 2022), gas storage status (Shang et al., 2020), shale

Yandy Scientific Press

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Received September 27, 2023; revised October 10, 2023; accepted October 15, 2023; available online October 18, 2023.

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gas preservation conditions (Gao et al., 2020), enrichment mechanism (Yan et al., 2003), and main controlling factors for high production (Zhao et al., 2023).

The Sichuan Basin and its periphery and the North American continent have similar basic geological conditions for shale gas formation and storage; however, they also have their own characteristics. For example, in the Sichuan Basin, (1) the shale formation system experienced the superimposed modification of multiple tectonic movements, which has led to stratigraphic uplift, denudation, folding deformation, and fracture/fault cutting, and the destruction of the stratigraphic pressure system (Guo, 2016); (2) the hydrocarbon source rocks are major detachment layers in the southern tectonic system as well as target layers for marine shale gas development (Zhu et al., 2019b); (3) the tectonism and preservation conditions are key criteria for shale gas enrichment and reservoir development in the context of multi-phase tectonic history and uplift denudation (Gou et al., 2022). As a result of the geological background of strong deformation and modification of the shale formation system, tectonics are a vital factor in the distribution and enrichment of shale gas reservoirs and are the core scientific issue that limits shale gas exploration in the complex tectonic area of Sichuan Basin.

2. Tectonic activity and shale deformation

Shale is the most widely distributed type of sedimentary rock in the shallow sublayer of the upper crust, which is not only of theoretical significance for the study of crustal tectonics but also of practical importance in the exploration of oil and gas resources (Liang et al., 2017). In structural geology, shale has page-like or sheet-like laminations. It is a kind of soft medium with strong rock plasticity and mobility, and is usually widely developed in foreland basins, passive continental margins, strike-slip basins, or extrusion tectonic environments as a detachment layer, which constitutes an important interface of shallow tectonic deformation systems within continents (Zhu et al., 2018). In oil and gas geology, shale layers are important cap layers, hydrocarbon producing layers and reservoirs, with the unconventional resources embedded in them constantly being emphasized (Gou et al., 2022).

Shales are characterized by the mechanical properties of rock such as low Young's modulus and high Poisson's ratio. In addition, they are fluid-rich and highly sensitive to metamorphic and deformation environmental factors, such as temperature, pressure, and tectonic stress (Zhu et al., 2018). As a result, the deformation/rheological behavior of shale in shallow sub-sedimentary rock layers of the Earth's crust is quite different from that of other inorganic rocks; even low tectonic stresses are sufficient to cause significant deformation and even the flow of shale layers at lower temperatures (Cheng et al., 2023).

Currently, the shale structural evolution and deformation mechanisms can be obtained in two ways. The first is to observe the shale in its natural deformation state at different scales to obtain its macro-micro structural deformation characteristics (Ma et al., 2015; Liang et al., 2017). The shale layers formed since the Early Paleozoic in the Upper Yangtze in the south of China have been involved in various retrograde thrust tectonic and slip tectonic systems during the later tectonic activities (Yan et al., 2003), and they have been subjected to strong interlayer shear, leading to different degrees of structural deformation. Liang et al. (2017) and Zhu et al. (2019b) observed and analyzed the field deformation characteristics of shale detachment layers. Some scholars have also attempted to describe brittle deformation and ductile deformation produced after the interlayer shear action of organic-rich shale to study the structural evolution mechanism (Zhu et al., 2018). Some fruitful research results have also been obtained on the rheological characteristics of shale. Li et al. (2021) studied the rheological structural characteristics of shale at different scales and their influencing factors using rock thin section and scanning electron microscopy.

In addition, artificial uniaxial, multi-axial, hightemperature, and high-pressure deformation tests were utilized to simulate the deformation process and elucidate the dynamic deformation mechanism (Cheng et al., 2021, 2022). Cheng et al. (2023) used the high-temperature and high-pressure triaxial experimental technique to simulate the structural modification process of shale during the period of tectonic deformation, and at the same time, obtained some rock mechanical parameters, such as compressive strength, Poisson's ratio, Young's modulus, shear strength, and brittle-ductile conversion. These findings comprised significant breakthroughs in revealing the process and mechanism of shale deformation from the aspect of simulation experiments.

3. Geologic factors affecting shale gas preservation

3.1 Tectonic activity

Unlike the favorable preservation conditions of North American shale gas, marine shale in southern China has been influenced by regional tectonic evolution, undergoing early burial and late rapid compression and uplift processes that played a crucial role in the preservation and enrichment of shale gas (Gao et al., 2017, 2019). The Sichuan Basin and its periphery, located in the middle-upper Yangtze region, experienced multiple stages of tectonic movements including the Caledonian, Hercynian-Indosinian, Yanshanian-Himalayan orogenies. These resulted in complex modification and diverse structural patterns (Yan et al., 2003). Among them, the current structural pattern primarily formed during the Yanshanian orogeny and further evolved during the Himalayan period. Tectonic evolution controls the opening of shale fractures, the enrichment of free gas, and the development of formation overpressure (Guo, 2016). Different tectonic evolution processes and intensities influences the occurrence state and gas content of shale gas, with late and low-intensity tectonic uplift being favorable for shale gas preservation.

3.1.1 Degree of shale fracturing

Structural compression and uplift can induce the generation of natural fractures in shale (Zhu et al., 2019a). Different intensities of tectonic activity lead to varying degrees of natural fracture development. Strong tectonic activity results in extensive fracture development, forming permeable pathways for shale gas flow (Zhu et al., 2018) and leading to rapid gas loss and unfavorable shale gas preservation. On the other hand, moderate tectonic uplift can create microfractures (i.e., the reopening of fractures), increasing the storage space for shale gas and promoting the desorption of adsorbed gas into free gas, which has a constructive effect on shale gas preservation (Zhu et al., 2019a).

It is generally believed that areas near large faults or those with well-developed microfractures have relatively poor preservation conditions for shale gas reservoirs. For example, in the Jiaoshiba shale gas reservoir, wells in the area with developed fractures generally have lower natural gas production compared to wells located away from faults (Gou et al., 2022). Large regional faults, due to their multiple phased and long-term activities, have well-developed microfractures and are influenced by atmospheric water infiltration, resulting in relatively poor shale gas preservation conditions in the surrounding areas. In regions where large fractures are underdeveloped or absent, the expulsion of hydrocarbons from shale is hindered, resulting in less hydrocarbon expulsion and more residual hydrocarbons present in shale, overall creating more favorable conditions for shale gas preservation and accumulation (Guo, 2016).

3.1.2 Tectonic evolution controls the shale gas content

Structural activities remodel the geological structure pattern, thereby controlling the dispersion of shale gas and resulting in significant differences in the shale gas content. Therefore, the intensity of post-structural movements is an important factor affecting the destruction and loss of shale gas reservoirs. Moderate uplift can maintain overpressure, reopen microfractures formed during early hydrocarbon generation and promote the desorption of adsorbed gas, thus playing a constructive role (Guo, 2016). The relationship between the uplift time, uplift magnitude, and shale gas content of Wufeng-Longmaxi shales in the Sichuan Basin and its periphery indicates a clear negative correlation between gas content and uplift time and magnitude (Gao et al., 2017, 2019). In other words, the earlier and greater the uplift of shale, the longer the duration and larger the amount of shale gas loss, which is unfavorable for shale gas preservation. Conversely, the later the uplift time and the smaller the uplift magnitude, the better the preservation conditions for shale gas, which is conducive to shale gas enrichment and high production (Gao et al., 2017, 2019).

3.2 Stratigraphic confinement

The confinement of shale gas primarily includes top and bottom sealing and self-sealing. The conditions of top and bottom sealing of the shale gas layer, as well as the self-sealing ability of shale, determine whether shale gas can be preserved and enriched within the shale gas reservoir during the early primary hydrocarbon generation stage (Gao et al., 2017, 2019). High-quality top and bottom sealing and favorable self-sealing conditions of shale gas can create a fluid trapping system with the gas-bearing shale interval, inhibiting the diffusion of shale gas and promoting its preservation. The self-sealing ability of shale is primarily related to parameters such as lithology, reservoir type, physical characteristics, and microsealing capacity. Top and bottom sealing respectively refer to the overlying and underlying formations directly in contact with the gas-bearing shale interval (Gou et al., 2022). The contact relationship between the top and bottom sealing and the shale gas reservoir, as well as the characteristics of the top and bottom sealing (such as lithology, thickness, lateral continuity, physical properties, and breakthrough pressure), are crucial for the preservation conditions of gas-bearing shale, which determine whether shale gas can be conserved and enriched within the shale reservoir during the early primary hydrocarbon generation stage. The top and bottom sealing mechanism for shale gas involves lithology and physical sealing, where tight lithology, shale thickness and micro-pore structure are the key controlling factors for top and bottom sealing. The self-sealing mechanism of shale gas involves capillary force and intermolecular forces, which are controlled by pore connectivity and methane adsorption (Zhu et al., 2019b).

3.3 Dissipation effect

Preservation is a dynamic process of continuous loss and relative sealing of shale gas. Sm-Nd isotope dating and fluid inclusion studies of veins in the Wufeng-Longmaxi shales revealed that the shales experienced a moderate to high overpressure gas evolution stage due to kerogen and liquid hydrocarbon thermal cracking into dry gas during the maximum burial depth stage (Gao et al., 2017, 2020). During the Yanshanian-Himalayan tectonic uplift, the shale gas reservoir underwent a dynamic evolution of cooling, depressurization, and gas loss (Gao et al., 2019). Simulation calculations were performed to investigate the evolution of shale gas content in the Wufeng-Longmaxi shales during the geological history of JY1 well in the Fuling shale gas field and that of PY1 well in the Pengshui shale gas field. The amount of shale gas loss during the uplift process was approximately 15.3%-16.9% of the total gas content during the maximum burial stage for well JY1, and it was approximately 27.3%-35.3% for PY1 well (Guo, 2016; Gao et al., 2017, 2019).

4. Structural evolution of shales based on natural geologic samples

4.1 Microstructural response to shale deformation

Through the analysis of the location, geometry, composition, connectivity, and formation mechanism of different pore types of shale, the deformation behavior and process can be observed and identified, and the macro- and microscale discrimination markers of deformed shale can be established (Liang et al., 2017; Li et al., 2020; Yang et al., 2022). In this study, the evolution characteristics and deformation modes of organic matter (OM) pores, interparticle (interP) pores, and intraparticle (intraP) pores and microfractures under tectonic deformation were examined (Fig. 1). These mainly include the shape and orientation change of OM pores (elongating,



Fig. 1. (a) Schematic model for the deformed evolution of pore structures from different parts of shale deformation structure (modified from Li et al. (2021)). Considering the characteristics of pore structural evolution, porosity and permeability, the shale reservoir in the shale deformation structure can be classified into five levels. Level (1): the undeformed shale. Level (2): the fold limb far from the fold core. Level (3): the fold limb near the fold core. Level (4): the fold core and the area close to the ductile shear zone. Level (5): the ductile shear zone. (b) Schematic diagram of the microstructural deformation characteristics of various pore types (modified from Li et al. (2021)). (c) SEM image for organic lamellar structures of the faulted shale sample (modified from Li et al. (2022)). (d) SEM image for quartz nanospheres of the faulted shale sample (modified from Li et al. (2022)).

relaxing, bending, etc.), extrusion and crushing of rigid particles, folding of clay layer or formation of high-angle fracture, dissolution, and closed pore extension, etc. (Fig. 1(b)). All pore types become interconnected during shale deformation and form microfractures ranging from a few nanometers to tens of microns, which is the main reason for the enhanced shale connectivity caused by structural deformation (Li et al., 2021).

Organic lamellar structures were observed inside the fault (Fig. 1(c)), along with inorganic nanoparticles (Fig. 1(d), indicated by white arrow), which comprise the unique microstructure at the fault site (Zhu et al., 2018; Li et al., 2021). Organic matter is in an amorphous state at the initial stage of formation and gradually begins to orient under shear action (Li et al., 2022). The lamellae of organic matter are parallel to the shear plane, and they are stretched and extended. As a result, the structure of organic matter becomes looser, the pore size increases (Fig. 1(c), OM pores are indicated by white arrows and dashed circles), and they are gradually connected with each other, forming a more effective microfracture network. Inorganic nanoparticles are produced by mechanical friction and thermal action under shear action and rearranged into flowing particles to relieve the local stress accumulation (Li et al., 2021). The generation of nanoparticles creates a rigid framework that preserves and increases the number of pores and enlarges the storage space for shale gas (Li et al., 2022). The presence of organic lamellar structures and inorganic nanoparticles in the fault provides more reservoir space for shale gas in the fault.

4.2 Reservoir physical response to shale deformation

With increasing deformation, micropores gradually become less abundant while the number of macropores increases (Liang et al., 2017; Li et al., 2021). The proportion of micropores in the fold core shale samples is the lowest at only 1/3 of that in the undeformed shale, while the proportion of macropores is as high as 1.2 times that in the undeformed shale (Li et al., 2022). The porosity and permeability of shales with strong deformation (ductile shear zone, fold core) are much higher than that of the weakly deformed (fold limb) and undeformed shale samples, and these characteristics have significant effects on the storage and migration of shale gas (Zhu et al., 2018). Under the same porosity conditions, the permeability of deformed shale is much higher than that of undeformed shale, and stronger deformation corresponds to higher permeability (Zhu et al., 2019a). This is consistent with the results obtained by laboratory deformation simulation experiments (Cheng et al., 2023). The fold core permeability is generally higher than the limb permeability, potentially exceeding it by 30 times. The different ductile deformation positions were divided into five levels, and the corresponding reservoir quality was evaluated. These results can provide data support for the evaluation of tectonic factors of shale gas. They also indicate that sites with strong ductile deformation should be the focus of attention.



Shale deformed under low-confining pressure showing **high-intensity brittle deformation** s

Shale deformed under moderate-confining pressure
showing moderate-intensity brittle deformationShale deformed under high-confining pressure
showing ductile deformation

Fig. 2. Deformation characteristics of experimentally deformed shale with varying mechanisms and intensities (modified from Cheng et al. (2023)).

5. Structural evolution of shales based on high-temperature and high-pressure simulation experiment

5.1 Deformation structures

Through shale deformation experiments, it was revealed that the deformation patterns and structures experience gradual evolutions with increasing confining pressure and temperature, and that the confining pressure plays a more important role than temperature in this process (Li et al., 2021, 2022; Cheng et al., 2023). Experimentally deformed shale samples of low confining pressure usually exhibited intense brittle deformation with apparent strain expansion characteristics. Two broad fracturing zones intersect in a conjugate shape, intensely breaking shale into fragments and particles (Fig. 2(a)). Within these strong deformation zones, dense secondary fractures develop; shale experiences intense fragmentation (Fig. 2(d)), and the shale and mineral fragments usually show angular and subangular shapes (Fig. 2(g)), indicating a brittle deformation

mechanism (Cheng et al., 2023).

With increasing confining pressure, brittle deformation features gradually weaken (Zhu et al., 2018; Cheng et al., 2023). The number of main fractures and the width of fracturing zones gradually diminish (Fig. 2(b)); meanwhile, the strain gradually concentrates in a limited area near the main fracture planes (Figs. 2(e) and 2(h)). Shale fragments within the fracturing zones mainly show sub-angular and sub-round shapes, indicating an increased roundness from those of low confining pressure. Some of the fragments display directional arrangement along their long axis. These evolution characteristics suggest the weakening of brittle tensile fracturing and the enhancement of compressive-shear fracturing (Zhu et al., 2019a; Cheng et al., 2023).

The development of fractures was significantly reduced when the confining pressure reached its threshold values (75-100 MPa). The further deformation of deformed shale under high confining pressure mainly concentrated in areas near individual main fracture planes where shale component bands show crumpled morphology (Fig. 2(c)). There is usually noticeable displacement along the main fracture planes. Under electron microscope, the secondary fractures are not developed within the strong deformation zones (Fig. 2(f)), and the main deformation mechanisms change into bending deformation and inter-layer sliding of clay minerals and the rheological deformation of shale matrixes (Fig. 2(i)). In this process, brittle mineral particles experience fine fragmentation and strain rounding, and the formed lenticular and ellipsoidal fragments rearrange along the rheological deformation direction (Cheng et al., 2023). As a result, shale samples display crumpled structures at macro- and mesoscales.

5.2 Deformation mechanism

Furthermore, we observed and summarized the microdeformation structures of shale components within the strong deformation bands of brittle and ductile deformed shale samples. It was found that the overall deformation patterns and structures of shale with increasing confining pressure were the comprehensive results of the differential deformation evolutions of different shale components at the microscale (Cheng et al., 2023). The deformation mechanisms of shale components experienced a transition from brittle mechanisms to ductile ones; different components in shale require varying confining pressure conditions for brittle-ductile transition (Li et al., 2021; Cheng et al., 2023). In general, clay minerals and organic matter are the components in shale that first undergo brittle-ductile transition with increasing temperature and confining pressure, which exhibited bending deformation and strain adjustment under a confining pressure of 25 MPa. Mica showed plastic deformation under a confining pressure of 75-100 MPa. As for minerals with high brittleness, such as quartz, feldspar, dolomite, and calcite, they mainly experienced brittle deformation. With increasing confining pressure, the brittle deformation mechanisms of brittle minerals gradually transitioned from simple mechanical fracturing (Figs. 2(g) and 2(h)) to 'intra-granular shear deformation, particle rotation, stress rounding' induced by the solid-state rheological deformation of shale matrixes (Fig. 2(i)).

In summary, we can conclude that with increasing confining pressure, the deformation pattern of experimentally deformed shale (mainly manifested as the combining form of main fracture planes) gradually transitioned from densely conjugated to sparsely conjugated and then to monoclinic pattern. The fracturing zones gradually narrowed down, and the deformation features within the strong deformation zones and near the main fracture planes changed from tensile brittle fracturing to compressive-shear brittle fracturing and subsequent rounding and directionally arranging of shale fragments (Cheng et al., 2023). In this process, the deformation mechanisms of shale components transitioned from brittle to ductile.

Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (No. 42102186) and the SINOPEC Key Laboratory of Geology and Resources in Deep Stratum (No. 33550000-22-ZC0613-0240).

Conflict of interest

The authors declare no competing interest.

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