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Research on tectonic control of late escape of Longmaxi formation shale gas in northeastern Chongqing

In northeastern Chongqing, the Longmaxi formation shale has experienced several tectonic uplifts of various scales during the geological evolution, hence forming shale gas enrichment zones featuring thrust fold belt. Based on field geology, drilling core characteristics observation and physical simulation experiment, and combined with methane isothermal adsorption experiment, this paper carries out a quantitative analysis on the variation rules of gas content during the late uplift of thrust belt in northeastern Chongqing, and discusses escape modes of Longmaxi formation shale gas by stages. The results show that the reservoir experiences two major tectonic uplifts after hydrocarbon generation ends, during which the changes in adsorption capacity are mainly affected by the combination of temperature and pressure. The content of adsorbed gas in the reservoir slightly increases, and the content of free gas decreases rapidly, thus the total gas content decreases. The simplified geological model and simulation restoration of tectonic burial history in northeast Chongqing are constructed. The shale remodelling in the study area is divided into four stages. The initial tectonic uplift amplitude was about 1,400m, and the total gas content was reduced; the second uplift amplitude was about 1,100m, and the total gas content was further reduced.

Keywords: *Shale gas 1, gas escape 2, gas content 3, Longmaxi formation 4, northeast Chongqing 5.*

1. Introduction

Shale gas is a hydrocarbon gas containing methane as the main component in gray-black carbonaceous shale in the dissolved, free or adsorbed states, which is an important unconventional natural gas resource[1-3]. Shale gas generation must first meet the needs of the surface adsorption of organic matter and rock. When the amount of adsorbed gas and dissolved gas reaches saturation, the rich natural gas migrates and accumulates in a free state[4-5]. According to the researches by Curtis[5], Mavor[6], Li Xinjing et al[7], and Nie Haikuan et al.[8], the content of adsorbed shale gas in the major shale gas producing areas in North America can account for 40%~85% of the total shale gas content. Exploration practice also shows that the production life of shale gas wells is usually relatively long, some may be as long as 30 years, and the declining rate of production is generally less than 5% (mostly 2%~3%). Many studies suggest that the reason for the longer and stable production period of shale gas well is closely related to the content of adsorbed gas in the reservoir, as the natural gas produced in the later stage of shale gas mainly comes from the adsorbed gas in the matrix[9-10]. Therefore, adsorption is one of the most important ways to preserve natural gas in shale reservoirs, and adsorption gas analysis is an important mechanism of shale stable production.

At present, the North American region in the Western Hemisphere and China in the Eastern Hemisphere are the two major shale gas business development bases in the world [11-[12]. Compared to North America, marine shale in southern China has undergone multiple stages of uplift and denudation. The study on the reconstruction and preservation conditions of late stage structure is more important for exploration and development of marine shale gas in southern China [13-15]. The post-deposition Longmaxi formation in northeastern Chongqing features “rapid settlement in the initial stage, small rise in the early stage, relative stability in short term and strong transformation in the latter stage” [16-17]. The uplift and erosion in the later stage lead to the changes in the temperature and pressure environment of the Longmaxi formation shale, and the content of free gas and adsorbed gas changes dynamically during the transformation process [18]. Therefore, to explore the changes of reservoir adsorption can get us to know the reason for the gas content loss during the whole evolution

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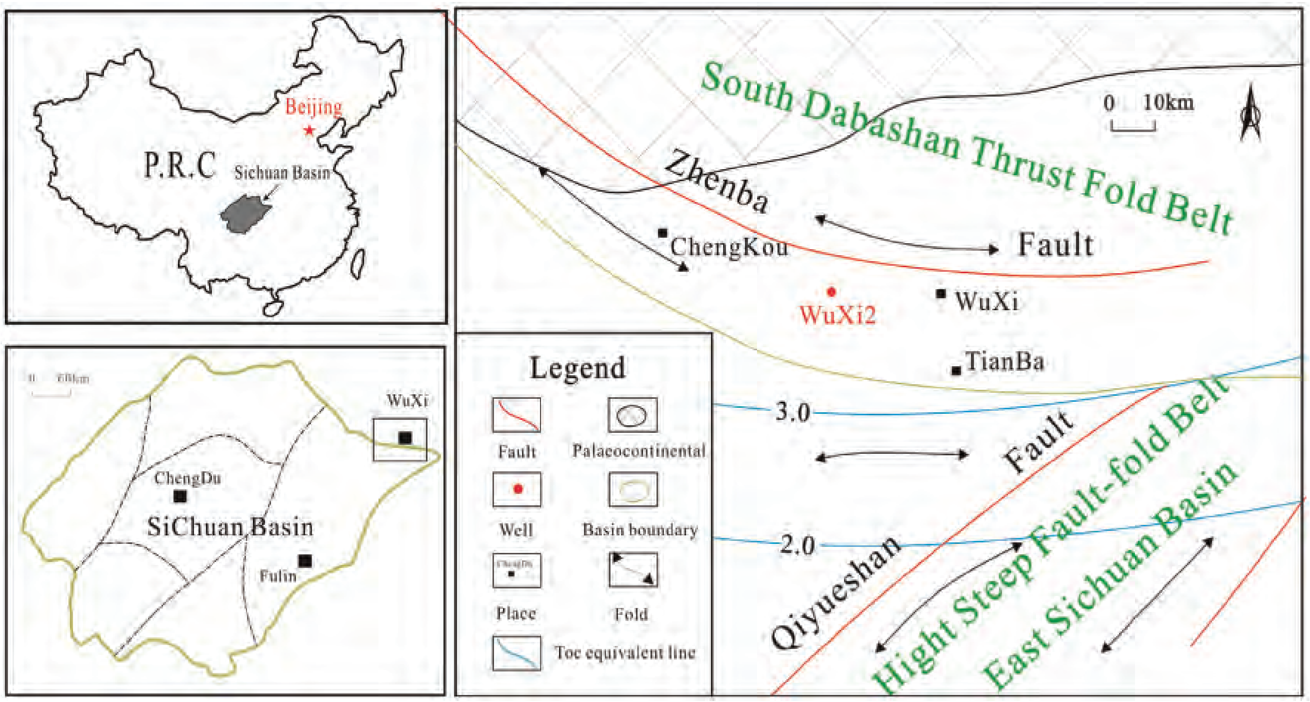


Fig.1 Geological structure and TOC contour of Longmaxi formation in the northeastern Chongqing

process of Longmaxi formation shale [19], which is the key to evaluate the gas content of the lower Paleozoic shale in the study area and to predict the favourable shale gas exploration zone.

Aiming at the strong shale gas escape of the Longmaxi formation in the later stage in northeastern Chongqing, this paper obtains the adsorption parameters of Longmaxi shale under different temperature and pressure conditions in northeastern Chongqing, discusses the variation rules of gas content in reservoirs during the structural uplift and denudation of the Longmaxi shale, and defines the influencing factors of late escape of shale gas in Longmaxi formation, through field structure measurement, drilling core and field outcrop sample collection and based on shale isothermal adsorption experiments, with a view to providing a scientific basis for the exploration and development of shale gas in the Longmaxi formation in this area.

2. Geological situation

Located at the junction of the northern margin of the middle Yangtze plate and the southern margin of South Dabashan, the northeastern Chongqing area belongs to the complex tectonic zone formed by multiple tectonic belts. The tectonic subdivision belongs to the South Dabashan thrust fold zone, consisting of a series of arc-shaped thrust faults and arc-shaped folds. The study area has experienced three stages of motion superposition, including early extension, mid-term settlement and late extrusion. Since the Indosinian movement, the study area belongs to a strong transformation zone of tectonic movements. Nowadays, the tectonic stress is complex, the southeastern direction shows a trough-like fold tectonic style, and the northwest direction shows a separated block-type fold tectonic style. In the study area, the arc anticlines are often associated with large thrust groups, while the syncline is relatively intact without fault development [20-21]. The sedimentary basement in northeastern Chongqing is composed of Banxi Group epimetamorphic rocks of the presinian system. Besides lacking devonian, carboniferous, and third lines, there has always been stratigraphic deposition from the Sinian to the Quaternary in the upper layer (Fig.1).

Longmaxi formation in the study area is a set of black carbonaceous shale with rich organic matter, including the black, greyish-green and gray carbonaceous shale, siliceous shale and sandy shale with rich content of graptolite fossils that are deposed in the shelf environment. Silurian source rock is the most widely distributed in the Lower Silurian Longmaxi formation shale (mudstone) [22-24]. Sedimentary facies of the Rhuddanian Longmaxi formation in the northeastern Chongqing area are dominated by the deep-water shelf facies, with the water depth becoming gradually shallower from south to north. A small range of sandy delta facies can be seen in the area south to the Northeast Chongqing. Controlled by the depositional environment, the Longmaxi formation in northeastern Chongqing is 160 m ~250m thick, and the lower black carbonaceous shale is 25 m ~110m thick. In most areas, the thickness of carbon shale is more than 50m. The TOC value is 2.18 % ~3.48 %, with TOC content in most areas greater than 2.0 % (Fig.1).

3. Experimental samples and testing methods

WX2 Well Longmaxi formation black organic-rich shale core was sampled for the present research. The isothermal adsorption experiment was carried out in the Experimental Center of Guizhou Provincial Bureau of Coal Geology, and the volumetric method was adopted for static adsorption. According to the changes of pressure, volume and temperature of the shale adsorption medium before and after adsorption, the adsorption process of shale was simulated, and the adsorption gas content under different pressures was calculated, by the mass balance equation, static gas balance and pressure measurement.

The experiment was carried out with temperature controlled at <130°C and pressure controlled at <40MPa. The adsorbed gas was methane and helium, with purity higher than 99.9%. Other materials, including moisture balance salt and distilled water, were in conformity with the corresponding test standards. Before the experiment began, the sample was broken into 60~80 mesh, and then the volume of the gas adsorbed by the rock sample under standard conditions was then tested at different temperatures and pressures.

4. Experimental results

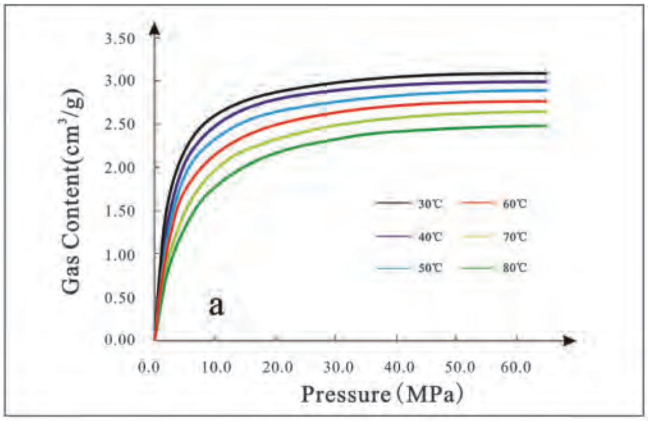
According to the isothermal adsorption experiment results of Longmaxi formation shale in eastern Chongqing area, shale gas adsorption capacity can be described with Langmuir equation under certain conditions of temperature and adsorption medium:

$$V = V_L * P/(P_L + P) \quad ... \quad (1)$$

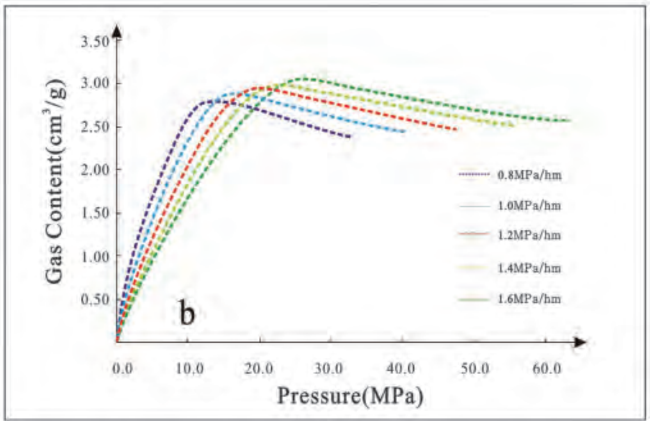
where, V is the adsorption capacity under a pressure of P; V_L is the volume of Langmuir; P_L is the pressure of Langmuir.

V_L can reflect the maximum adsorption capacity of shale; P_L is the ratio of the desorption rate constant to the adsorption constant, indicating that the pressure at which the adsorption capacity of shale is half of the maximum adsorption capacity, that is when $V = V_L/2$, then $P = P_L$. V_L and P_L determine the adsorption capacity of shale under different temperature and pressure. Table 1 shows V_L and P_L calculated from the shale isothermal adsorption experiments.

The previous studies on the paleo geothermal field in northeast Chongqing show that geothermal gradient is 2.5°C/hm ~2.8°C/hm after Silurian deposition. Combining test features of the experimental instruments, this paper adopts a geothermal gradient of 2°C/hm and selects different geothermal gradients, including 0.8MPa/hm, 1.0MPa/hm, 1.2MPa/hm, 1.4MPa/hm and1.6MPa/hm, to simulate different effects of different uplift scales on adsorption capacity of shale in Longmaxi formation in different periods. The shale isothermal sorption curve measured by the laboratory is shown in Fig.2(a). The parameters of V_L and P_L obtained by fitting the experimental data are shown in Table 2. Given the actual geothermal and pressure gradients, the temperature and pressure at different depths can be calculated, and then the adsorption capacity of shale in Longmaxi formation under various burial depths is calculated according to the Langmuir equation (Equation 1). In this way, the changes of shale adsorption capacity at different depths (temperature and pressure) during tectonic uplift are retrieved. Before starting the study, due to the experimental apparatus limitations, the partial boundary conditions were set under the premise of not affecting the experimental science as much as possible. Assuming that the maximum burial depth of the shale in the Longmaxi formation shale is 4km and the maximum lift is 2.5km, the surface temperature and pressure are both 0, without considering the influences of the temperature of the constant temperature zone in each geological history. According to Eq(1), adsorption capacity under geothermal gradient is 2°C/hm and different pressure gradients are calculated, the results are shown in Fig.2(b).



(a) Experimental data



(b) Temperature gradient 2°C/hm, different pressure gradient

Fig.2 Adsorption characteristics of Longmaxi formation shale under different conditions

5. Discussions

5.1 Tectonic evolution characteristics of the Longmaxi formation shale gas reservoirs in the northeastern Chongqing

The research on the burial history of Longmaxi formation and the hydrocarbon generation history of shale in the

TABLE 1: LANGMUIR PARAMETERS OBTAINED FROM THE ISOTHERMAL ADSORPTION EXPERIMENT OF WX2 WELL LANGMAXI FORMATION SHALE

Temperature/°C	V_L/ ($cm^3 \cdot g^{-1}$)	P_L/MPa
30	3.20	2.31
40	3.12	2.48
50	3.03	2.94
60	2.94	3.55
70	2.85	4.56
80	2.80	5.80

northeast Chongqing shows that there is a big difference between the hydrocarbon generation and evolution of dark shale in Longmaxi formation of northeastern Chongqing and that of the typical North American shale gas development zones, such as Barnett, Woodford, etc. The North American shale is characterized by low maturity, relatively small depth, simple tectonic activity, gentle formation and complete reservoir. The long period of burial and multiple rapid uplift processes that Longmaxi formation shale in northeastern Chongqing has undergone, the location in the southern margin of Dabashan fold thrust belt, and the development of thrust nappe tectonic belt have made the shale distribution of the Longmaxi formation to feature favourable areas with grid distributions. In order to understand the relationship between tectonic evolution and shale gas in northeastern Chongqing, this paper chooses to pass through the AA' section of Well WX2 and combines with the theory of tectonic geology and continuous mechanics medium theory, to establish a simplified geological model for AA' section of shale gas reservoir with the help of 2D Move (Fig.3). In this model, the formation and evolution of the Longmaxi shale gas reservoir can be divided into the following four stages:

Stage one

Slow settlement, the compacted shale reservoir enters the oil window stage. Longmaxi shale organic matter features larger pore diameter, relatively stable structure, and the nearly horizontal strata. As the burial depth gradually increased, and the temperature of geotemperature rose, organic matter became more and more mature, and entered the oil window stage.

Stage two

The continuous deep burial brings about the maximum burial depth stage. After the shale entered the oil window stage, the strata settlement slowed down or rose slightly, and gradually reached the maximum burial depth, with the depth of some regions more than 5,000m. Rapid deep burial caused the rapid decrease of pores in shale, the permeability of reservoir reduced, and the shale thermal maturity evolved quickly. The oil and gas generated in the early stage were split to generate gas, and as time went on, the hydrocarbon generation process gradually ended.

Stage three

Initial uplift stage. In the early Cretaceous, subducted by the Yangtze block moving from the south to the north and the Qinling Mountains orogenic belt, South Dabashan area entered thrust fold deformation period, bringing about a large scale thrust nappe and deep slipping and shearing effects. The strata underwent the initial large-scale uplift since their deposition. The duration of this uplift was about 15Ma and the area of this formation was about 1,400m. The thrust nappe fault was formed in Wuxi area, and the region was dominated by fold structure. Large-scale secondary microfractures were formed on both sides of the fault and micro-fractures formed

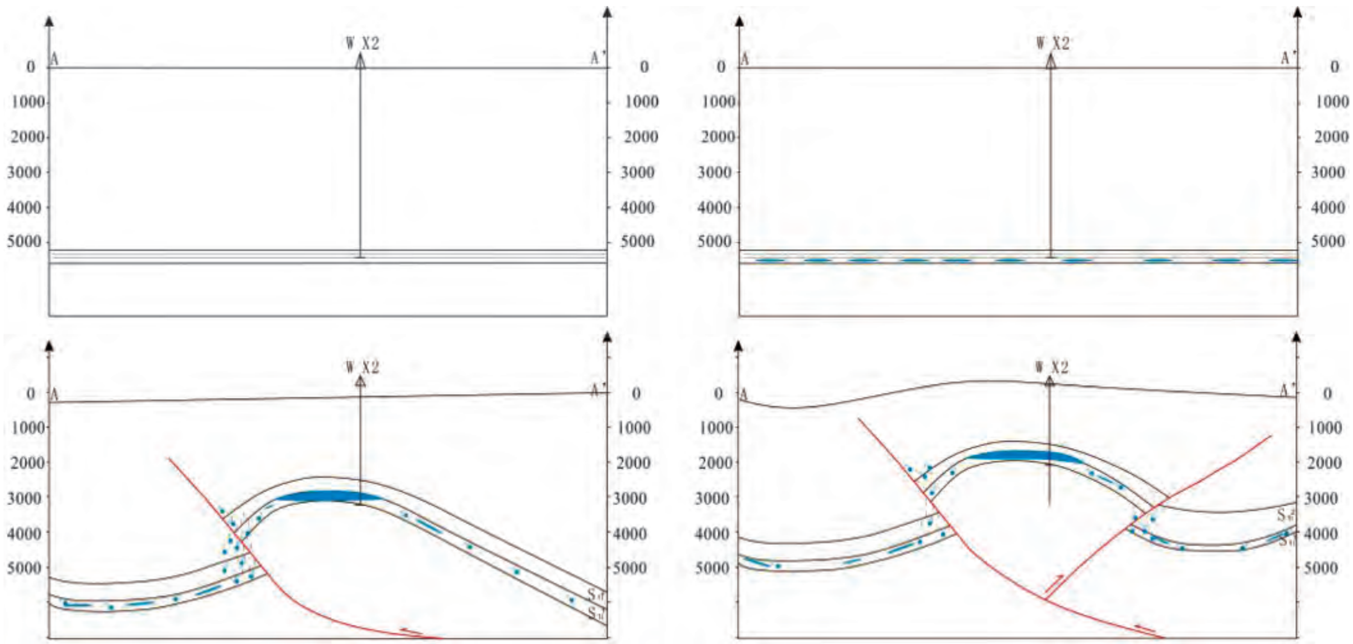


Fig.3 Tectonic evolution model of shale gas reservoir in northeastern Chongqing

by the shale gas fault zone escaped upward. The hydrocarbon concentration was inversely proportional to the fault distance. The anticline structure was gradually formed in this tectonic stage in Wuxi, and the shale gas generated at early stage migrated upward along the dip of the strata and converged at the nucleus of the anticline.

Stage four

The second uplift stage. After the strong initial uplift, northeast Chongqing was in a long period of gas reservoir adjustment stage until the late Himalayas. At about 20Ma, the second extrusion and uplift occurred to the Yangtze block and the southern Qinling Mountains orogenic belt. Based on the original thrust nappe structure, the extrusion superimposed more secondary faults. With the formation of more cracks with the fault zone, gas reservoirs suffered secondary migration. The tectonic transformation at this stage enhanced the connectivity of fractures in the shale. Shale gas escape occurred near the fault, and the gas reservoirs at the fold-modification site migrated to the nucleus of the anticline, creating the local "dessert zone" under the control of the structure in northeastern Chongqing.

5.2 Comprehensive influences of temperature and pressure on gas content

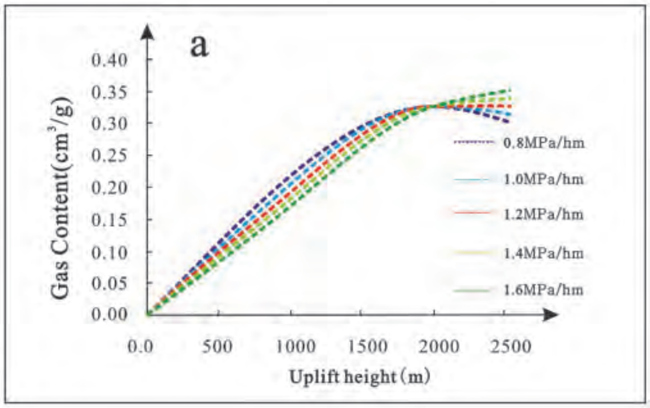
The effect of temperature on shale gas adsorption is negative. When the other conditions are constant, the adsorption capacity decreases with the increase of temperature, mainly because the adsorption is an exothermic reaction, and the temperature has an activation effect on methane desorption. The higher the temperature is, the stronger will be the activity of the gas molecules, the higher is the content of the free gas, and less is the adsorption gas. The adsorption capacity decreases with the increase of temperature (Fig.2a). It can be seen from the figure that the adsorption amount of methane increases rapidly with increasing pressure at different temperatures, and then the adsorption curve remains gentle and eventually tends to be stable. However, the higher the temperature is, the greater pressure will be for the adsorption capacity to reach the stable stage, and the smaller will be the maximum adsorption capacity.

When the geothermal gradient is 2°C/hm and the pressure gradient is 0.8MPa/hm, 1.0MPa/hm, 1.2MPa/hm, 1.4MPa/hm and 1.6MPa/hm, respectively, the maximum adsorption capacity of shale is 2.70cm^3/g, 2.78cm^3/g, 2.84cm^3/g, 2.88cm^3/g and 2.92cm^3/g, respectively. Moreover, the higher the pressure gradient is, the lower will be the temperature required for shale adsorption to reach its maximum. As can be seen from Fig.2(b), when the geothermal gradient is 2 °C/hm and the pressure gradient is lower than or equal to 1.0 MPa/hm, the shale adsorption capacity reaches the maximum at 40°C, indicating that when the pressure gradient is small (lower than or equal to 1.0 MPa/hm), the increasing shale adsorption due to the decrease of temperature and the decreasing shale adsorption caused by the decrease of pressure firstly increase and then decrease with the decrease of burial depth; when the geothermal gradient is 2°C/hm and the pressure gradient is greater than 1.0 MPa/hm, the shale adsorption capacity reaches the maximum at 30°C, indicating that when the pressure gradient is large (higher than 1.0 MPa/hm) the increasing shale adsorption due to the decrease of temperature and the decreasing shale adsorption caused by the decrease of pressure always increase with the decrease of burial depth.

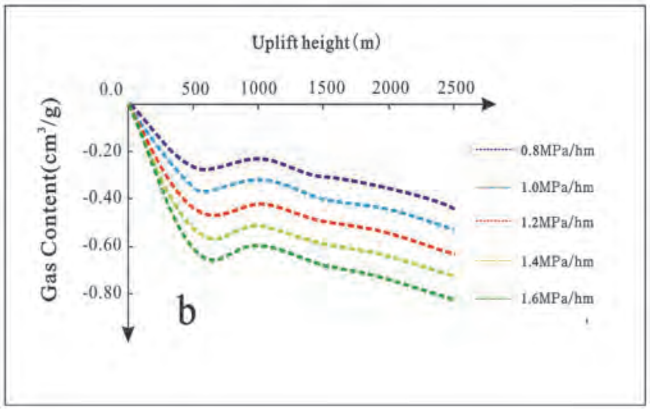
Therefore, the comprehensive effect of temperature and pressure in the process of tectonic uplift will have a great influence on the adsorption capacity of shale reservoir. Under different geothermal gradients and pressure gradients, the different variation rules of the adsorption capacity caused by the tectonic uplift are mainly attributed to the comprehensive effect of temperature and pressure, that is, there is an inversely proportional relationship between the changes in methane adsorption caused by temperature and pressure. When the strata are subjected to tectonic uplift at a later stage, the decrease in reservoir temperature leads to an increase in methane adsorption and a decrease in pressure results in a decrease in adsorption. When the change in adsorption capacity caused by temperature is more than that caused by pressure at a certain stage of uplift, the reservoir in this stage shows an increasing process of adsorption, otherwise, the reservoir shows a decreasing process of adsorption. At the same time, the changes in the adsorption capacity caused by the elastic changes of the nanoscale pores in the reservoir of the tectonic uplift cannot be ignored.

5.3 Comprehensive influences of temperature and pressure on gas content

The strength of the uplift has a certain effect on the adsorption capacity of shale reservoir. The relationship between the amount of stratigraphic uplift and the change in adsorption (geothermal gradient, 2°C/hm) can be calculated, according to adsorption capacity calculation curve and the simulated temperature and pressure conditions. As can be seen from Fig.4(a), during the uplift of the Longmaxi Formation shale, the content of methane adsorbed gas in shale gradually increases. When the uplift amplitude is less than 2,000m, the larger the pressure gradient is, the smaller the increase of adsorption gas content will be, indicating that the low pressure gradient during the shale uplift can easily lead to the rebound of the nanoscale pores in the shale, pore specific surface area increases rapidly, and the adsorption gas content increases more rapidly when the stratum pressure gradient is 0.8MPa/hm than 1.6MPa/hm. When the uplift amplitude is larger than 2,000m, the larger the pressure gradient is, the larger will be the increase of adsorption gas content, indicating that the low pressure gradient leads to limited increase of rebound in pore specific surface area of shale, and the adsorption gas content increases more rapidly when the stratum pressure gradient is 1.6MPa/hm than 0.8MPa/hm.



(a) Changes in the amount of adsorbed gas



(b) Changes in total gas content

Fig.4 Relationship between the uplift and gas content characteristics under different pressure gradients

It can be seen from, Fig.4(b), the gas content curve calculated under different pressure gradients that during the initial period of uplift (the uplift amplitude is less than 500m), the content of adsorbed gas slowly increases and the total content of gas decreases rapidly; when the uplift amplitude exceeds 1,000m, the total gas content will decrease slowly. Therefore, the intensity of tectonic uplift has a direct impact on the occurrence of shale gas in northeastern Chongqing. When the uplift is small, the increase of adsorbed gas in shale reservoirs is little, the amount of free gas escaped is little, the total gas content changes rapidly, but the total gas content in the reservoir is high; when the lift is large, the amount of adsorbed gas in shale reservoirs increases greatly, the amount of free gas escaped is massive, the total gas content tends to be stable, and the total gas content in reservoirs is low.

During the process of stratigraphic uplift, the temperature and pressure will change, and then the gas content of shale will change under the combined effect of

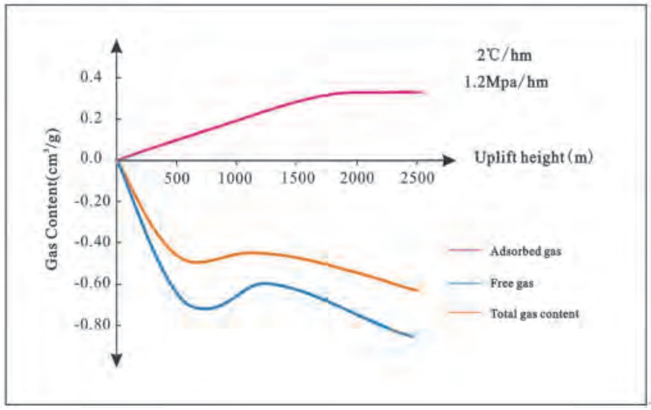


Fig.5 Relationship between tectonic uplift and changes in gas content characteristics

temperature and pressure. According to the previous analysis, the total gas content of shale is decreasing during the process of tectonic uplift (Fig.5). However, there is a significant difference between the reduction rate with different geothermal gradients and pressure gradients. When the geothermal gradient is very low and the pressure gradient is high, shale desorbs shale gas at a lower rate.

5.4 Comprehensive influences of temperature and pressure on gas content

The Petromod software is used to restore the burial history of WX2 well in northeastern Chongqing and draw the evolution map of the burial history. The stratum thickness and the lithology of each stratum used in the restoration process of Petromod are calculated according to the measured results of drilling. The model assumes that the formation is under continuous heat flux over time and is closely related to the thermal conductivity of the rock. The analysis of burial history shows that in the northeast of Chongqing, there appeared two hydrocarbon generations, one deep burial and two uplifts with different scales (Fig.6). In the initial stage of deposition, the strata of Longmaxi formation subsided at a

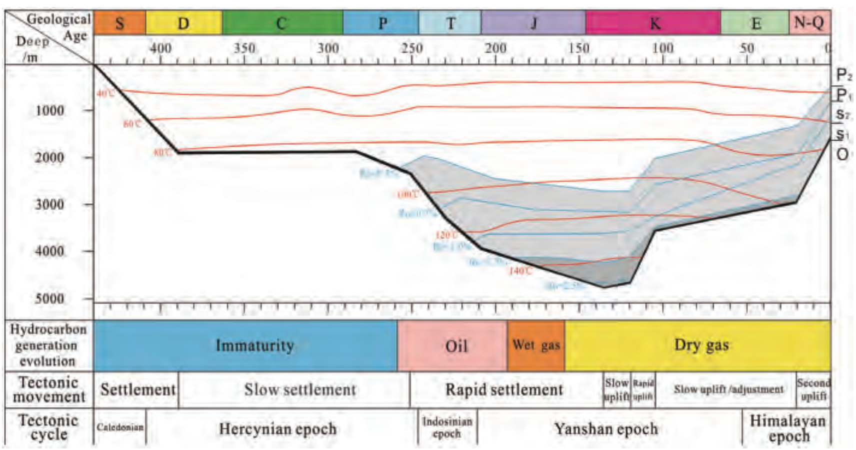


Fig.6 Recovery of the burial history of WX2 well Longmaxi formation in northeastern Chongqing

slow rate, followed by a short period of stabilization during the 390Ma~280Ma, then followed by a rapid settlement at an average rate of 0.019 km/Ma, which was a deep burial process of the Longmaxi formation. During this process, Ro of Longmaxi formation increased rapidly, reaching to 1.3%. Then in the moisture stage, the initial hydrocarbon generation occurred, and the maximum burial depth of Longmaxi formation reached 4,800m at 140Ma with a maturity of 2.52%. After reaching the maximum burial depth, the structure was relatively stable in a short period of time, and the oil and gas reservoirs have entered the adjustment phase after hydrocarbon generation. At the end of the adjustment phase at 120 Ma, the first rapid uplift occurred after the hydrocarbon generation in the Longmaxi formation with an average rate of 0.087km/Ma. The reservoir entered a phase with relatively stable structure at 105Ma, with the uplift rate slowing down, then came a transient transitional phase. The second uplift occurred in the Longmaxi formation at 20Ma with an average uplift rate of 0.075km/Ma, which was a large-scale uplifting event in the middle and later Himalayas and had a significant impact on the total gas content in the shale.

6. Conclusions

(1) The formation and evolution of the Longmaxi shale gas reservoir can be divided into 4 stages: slow settlement, the compacted shale reservoir enters the oil window; continuous deep burial, the oil and gas generated in the early stage are cracked and the hydrocarbon generation process gradually ends; initial uplift, shale gas migrates over a short distance and accumulates at favourable sites; second uplift and tectonic finalization, early shale gas reservoir is damaged and adjusted to finally form reservoir again.

(2) The effect of temperature on shale gas adsorption is negative. When the other conditions are constant, the adsorption capacity decreases with the increase of temperature. The higher the temperature is, the greater will be the pressure required to reach the stable stage of adsorption, and the smaller will be the maximum adsorption capacity. The effect of pressure on shale gas adsorption capacity is positive. When the other conditions are constant, the higher the pressure gradient is, the higher will be the adsorption capacity of shale, lower the temperature at which the maximum adsorption capacity is reached, and greater is the maximum adsorption capacity.

(3) The comprehensive effect of temperature and pressure in the process of tectonic uplift will have a great influence on the adsorption capacity of shale reservoir and there is an inversely proportional relationship between the changes in methane adsorption caused by temperature and pressure. When the strata are subjected to tectonic uplift at a later stage, the decrease in reservoir temperature leads to an increase in methane adsorption and a decrease in pressure results in a decrease in adsorption. When the change in adsorption capacity caused by temperature is more than that caused by pressure at a certain stage of uplift, the reservoir in this stage shows an increasing process of adsorption, otherwise, the reservoir shows a decreasing process of adsorption.

(4) In the first uplift of northeast Chongqing, the pressure gradient was 0.8MPa/hm, the temperature gradient was 4.1°C/hm, the uplift amplitude was about 1,300m, and the change in shale gas adsorption capacity was 0.36 cm^3/g; in the second uplift, the pressure gradient was 1.2MPa/hm, the temperature gradient was 2.6°C/hm, the uplift amplitude was about 1,500m, and the change in shale gas adsorption capacity was 0.39 cm^3/g; besides, the total gas content of Longmaxi formation decreased in both stages of uplift.

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