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Review Article

Lower Cambrian Organic-Rich Shales in Southern China: A Review of Gas-Bearing Property, Pore Structure, and Their Controlling Factors

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The Lower Cambrian shales are widely developed in southern China, with greater thicknesses and higher TOC contents. Although the shale gas resource potential has been suggested to be huge, the shale gas exploration and development is not satisfactory. At present, the gas-bearing property evaluation of the Lower Cambrian shale is still a hot spot of concern. According to previous works, this paper systematically summarizes the gas-bearing characteristics and controlling factors of the Lower Cambrian shales in southern China. The buried depth of Lower Cambrian shales mainly ranges from 3000 m to 6000 m, and the thickness of organic-rich shale intervals (TOC > 2%) varies from 20 m to 300 m. The TOC content and EqVRo value are generally up to 2%-10% and 2.5%-6.0%, respectively. The gas content of the Lower Cambrian shales in the Weiyuan-Qianwei block of the Sichuan Basin and the western Hubei area generally exceeds 2 m^3/t, and gas composition is dominated by CH_4. In southeastern Chongqing, northwestern Hunan, and northern Guizhou areas, the gas content of the Lower Cambrian shales is generally <2 m^3/t, and the N_2 content is generally >60%. In the Lower Yangtze region, the Lower Cambrian shale reservoirs basically contain no gas. Higher maturity, lower porosity, and less-no organic pores are suggested to be responsible for low gas contents and/or the predominate of N_2 in shale gas reservoirs. Strong tectonic deformation is an important factor leading to the massive gas loss from shale reservoirs, thus resulting in no gas or only a small amount of N_2 in the Lower Cambrian shales. In a word, the Lower Cambrian shale gas plays with low maturity and relatively stable tectonic condition, especially deep-ultradeep zones, may be the favorable targets for shale gas exploration.

1. Introduction

With the continuous growth of global oil and gas demand and the continuous decline of conventional oil and gas production, unconventional oil and gas with great resource potential has gradually become a new field, which has been highly valued by various countries and oil companies [1–7]. Total global production of shale gas in 2020 is 7688×10^8 m^3, which is mainly derived from North America, China, and Latin America. Among them, the United States is the main country of global shale gas development and production in 2020, with total shale gas yield of 7330×10^8 m^3[8]. The successful exploitation of shale gas in the United States has provided numerous valuable experiences for China.

China started shale gas exploration and development as early as 2010, and shale gas pilot tests for geological selection evaluation and development in different onshore areas had been conducted, revealing that China had abundant shale gas resources, especially marine shale gas [9, 10]. Since commercial exploitation of shale gas in China had been conducted in 2014 [8], the proven reserves and production of shale gas had exceeded 2×10^{12} m^3 and 200×10^8 m^3 in 2020, respectively [11]. At present, China ranks second in the world in terms of shale gas production.

The Lower Cambrian shales are important marine source rocks in southern China and are also key targets for shale gas exploration [12–18]. Such a shale succession is widely distributed, with greater thickness and total area up to $(30 - 50) \times 10^4$ km^2, and the predicted geological resource of shale gas is as high as 35.16×10^{12} m^3 (<

4500 m) [1, 19, 20]. However, the exploration and development of the Lower Cambrian shale gas in many blocks had not made substantial breakthroughs [21–24]. At the same time, the maturity of the Lower Cambrian shales is very high and at the overmature stage (equivalent vitrinite reflectance (EqVRo) > 3.0%-3.5%). Due to strong tectonic deformation, the gas content of the shale changes greatly and the influencing factors are complex [25–30]. Thus, these factors have hindered the process of exploration and development to some extents.

Shale gas belongs to a self-generated and self-storage unconventional natural gas [31, 32]. Organic matter (OM) enrichment is the material basis for shale gas generation. Shale has a certain porosity and nanopore network, which is the basic condition for shale gas accumulation. Preservation condition is another key factor affecting shale gas enrichment [33]. These factors are closely related to shale composition, thermal evolution, sedimentary facies, and other geochemical property, and they are controlled by geological conditions such as the intensity and mode of tectonic activity [34–42]. Gas-bearing property (gas content, composition, and occurrence form) of shale is the embodiment of comprehensive effect of these factors. Understanding the impact of these factors on the gas-bearing property of shale can help to screen the core blocks of shale gas and thus evaluate the exploration and development potential of studied gas shale reservoirs, which is more prominent for the Lower Cambrian shales in southern China.

In recent years, there have been many literatures on the Lower Cambrian shales in southern China and are mainly contributed by Chinese scholars (Figure 1). Among them, many publications are related to the evaluation, exploration, and development of the Lower Cambrian shale gas in southern China. Therefore, based on a large number of previous works, this paper systematically reviews the geochemical characteristics, current situation of shale gas exploration and development, and the gas-bearing characteristics of the Lower Cambrian shales in southern China and summarizes the development mechanism, the source of nonhydrocarbon gas, and the influence of tectonic deformation on the gas-bearing property of shale, thus providing a reference for further exploration and development for shale gas.

2. Geochemical and Gas-Bearing Characteristics of the Lower Cambrian Shales

2.1. Geochemical Characteristics. The Lower Cambrian shales in southern China are widely distributed across the whole Yangtze Platform. Organic-rich shale intervals are mainly distributed in the Sichuan Basin, western Hubei, Chongqing, northwestern Hunan, northern Guizhou, and southern Anhui areas, which are mainly deposited in the deepwater shelf facies (Figure 2). Affected by various factors such as sedimentary facies and structures, the lower Cambrian stratas are named differently in different regions (Figure 3). Qiongzhusi Formation in southwestern Sichuan, Niutitang Formation in northern Guizhou, southeastern Chongqing and northwestern Hunan, Shuijingtuo Formation in northeastern Chongqing and western Hubei, and Hetang Formation in southern Anhui are all equivalent. The thickness distribution of Lower Cambrian organic-rich shales has two centers, which are located in the Deyang-Anyue ancient rift trough in the Sichuan Basin and the southeastern margin of the Yangtze Platform, respectively. The former is controlled by the rift trough with thickness of 60 m-300 m, while the latter is controlled by sedimentary facies with thickness of 30 m-120 m [43–46]. These areas are also the main potential targets for the Lower Cambrian shale gas exploration and development currently.

In recent years, extensive geochemical studies have been carried out on the Lower Cambrian shales and some basic understandings are obtained [23, 24, 44, 51–58]. A brief overview of three aspects, including TOC, OM type, and maturity, can be concluded in the following.

The TOC content of the Lower Cambrian shales in southern China varies greatly, ranging from 0.1% to 15% (Table 1). In the southern part of the Sichuan Basin, the TOC content is generally in the range of 2.0% to 3.0% [12, 54, 57, 59]. In the Yangtze regions outside the Sichuan Basin, the TOC content of shales can reach as high as 5% to 10% [23, 24, 55, 58, 60–65]. TOC is one of the important indicators to evaluate the exploitation value of shale gas. At present, the lower limit of TOC for commercial shale gas development is generally 2.0% [66–69]. Therefore, the Lower Cambrian shales generally display greater shale gas potentials in terms of the TOC evaluation.

The organic macerals of the Lower Cambrian shales are mainly composed of sapropelite, and the parent materials are derived from lower aquatic organisms [12]. The $\delta^{13}C$ value of kerogen is -35.9‰–-29.2‰, with an average of -32.0‰ [23, 44, 57, 70, 71], so the OM type of the Lower Cambrian shales is mainly type I [72, 73].

In the whole Yangtze region, the Lower Cambrian shales have high maturity, with the EqVRo value ranging from 2.5% to 6.0% [30, 33, 56, 74–78]. The EqVRo value of the Lower Cambrian shales in the Upper-Middle Yangtze region mainly varies from 3.0% to 4.0%. In the Lower Yangtze region, the maturity is relatively higher, and the EqVRo value is mainly in the range of 3.5%-4.5% and even exceeds 4.5% in some areas (Table 1). The high maturity of the Lower Cambrian shales is closely related to their old age, large burial depth, and multiple thermal events [79]. According to shale gas data in the United States, shale gas reservoirs can also develop under high overmaturity conditions [80, 81], but the maturity of shale with commercial potential is generally limited within the EqVRo < 3.5% [75, 82].

2.2. Gas-Bearing Characteristics. At present, the number of wells drilled for the Lower Cambrian shale gas evaluation, exploration and development in southern China has reached 70-80 [23, 24, 79, 96–98]. Gas-bearing characteristics of the Lower Cambrian shales from representative wells are summarized in Table 2. In general, the gas-bearing property of the Lower Cambrian shales varies greatly. The shale reservoirs in most areas/blocks contain low gas content or are rich in N_2. Few shale gas wells in the Weiyuan-Qianwei block of the Sichuan Basin, the Yichang area of western Hubei and Chengkou area of northern Chongqing areas

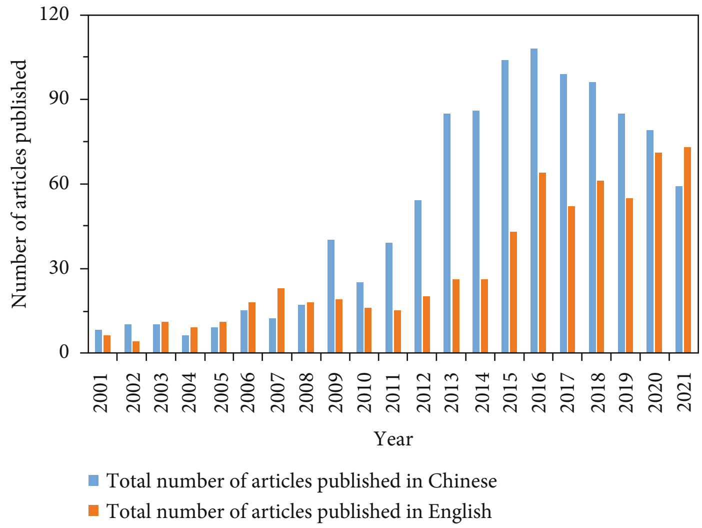

FIGURE 1: Statistical histogram of literatures related to the study of the Lower Cambrian shales and shale gas in China. The data of Chinese articles (blue column) comes from the "CNKI," the data of English articles (orange column) comes from the "ScienceDirect." The search methods are all through title, abstract, and keywords.

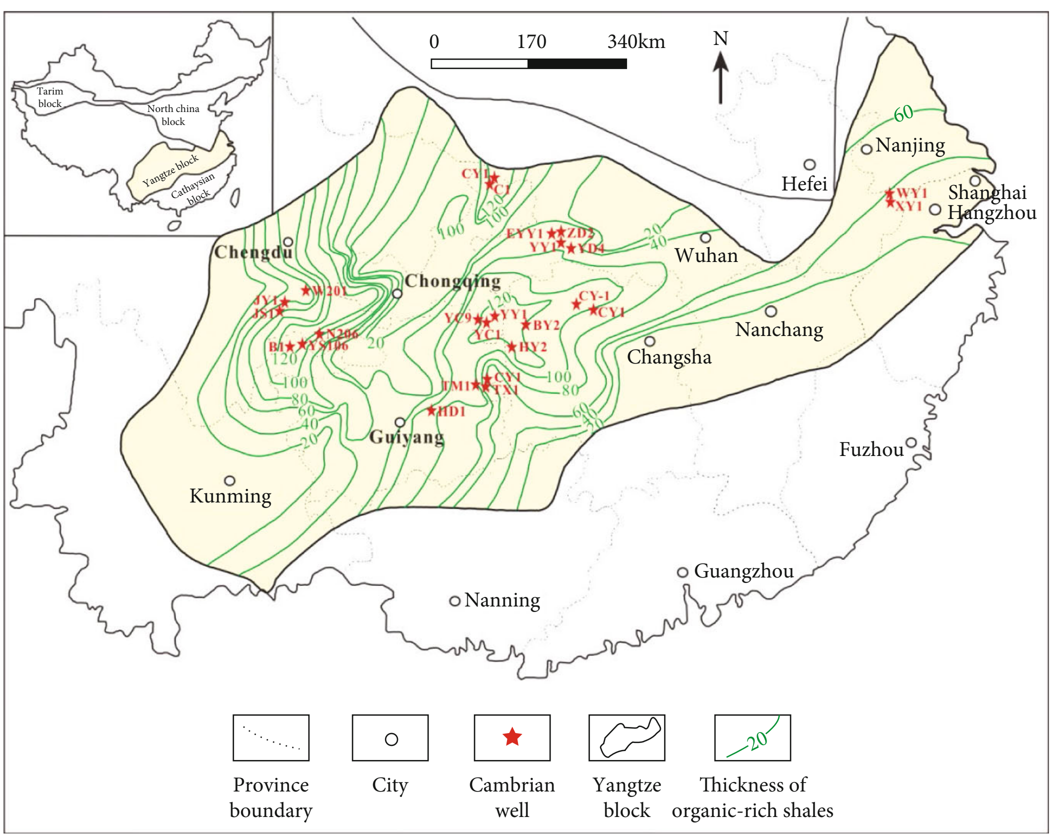

FIGURE 2: The distribution of the Lower Cambrian shale gas wells, thickness of organic-rich shales. CY-1: Ciye1 Well, CY1 in northern Hunan area: Changye1 Well (modified from Wu et al. [16]; Zhu et al. [45]; Wang et al. [47])

<table>
<tr><th rowspan="2">Strata</th><th colspan="7">Regions</th></tr>
<tr><th>Southwestern Sichuan</th><th>Northern Guizhou</th><th>Southeastern Chongqing</th><th>Northeastern Chongqing</th><th>Western Hubei</th><th>Northwestern Hunan</th><th>Southern Anhui</th></tr>
<tr><td rowspan="6">Lower Cambrian</td><td rowspan="2">Longwangmiao Fm.</td><td colspan="2">Qingxudong Fm.</td><td colspan="3">Shilongdong Fm.</td><td>Dachenling Fm.</td></tr>
<tr><td colspan="2" rowspan="2">Jindingshan Fm.</td><td colspan="3" rowspan="2">Tianheban Fm.</td><td rowspan="5">Hetang Fm.</td></tr>
<tr><td rowspan="2">Canglangpu Fm.</td></tr>
<tr><td colspan="2">Mingxinsi Fm.</td><td colspan="3">Shipai Fm.</td></tr>
<tr><td>Qiongzhusi Fm.</td><td colspan="2">Niutitang Fm.</td><td colspan="2">Shuijingtuo Fm.</td><td rowspan="2">Niutitang Fm.</td></tr>
<tr><td>Maidiping Fm.</td><td>Liuchapo Fm.</td><td></td><td></td><td>Yanjiahe Fm.</td></tr>
<tr><td colspan="8">Ediacaran</td></tr>
</table>

Figure 3: Stratigraphic division and correlation of the Lower Cambrian typical plays (modified from Zhu et al. [45]; Hu et al. [48]; Zhang et al. [49]; Zhao et al. [50]).

can yield industrial gas flow and the gas compositions are dominated by CH_4.

In the Weiyuan-Qianwei block of the Sichuan Basin, the gas-in-place (GIP) content of the Qiongzhusi Formation shales from the Well W201 ranges from 1.10 m^3/t to 3.51 m^3/t, with an average of 2.01 m^3/t, and initial test yield of 1.08×10^4 m^3/d. The Qiongzhusi Formation shales in the Well JY1 have a gas content of 1.02 m^3/t-4.68 m^3/t, with an average of 2.03 m^3/t, and the initial test yield of 8.60×10^4 m^3/d [96]. In the Yichang area, the GIP content of the Well YY1 varies from 0.58 m^3/t to 5.48 m^3/t, with an average of 2.05 m^3/t. The thickness of the organic-rich shale intervals with the GIP content greater than 2 m^3/t is 35 m, and the initial yield of horizontal well fracturing section is 6.02×10^4 m^3/d. The GIP content of the Well EYY1 is 0.32 m^3/t-4.48 m^3/t, with an average of 2.3 m^3/t, and the initial yield of horizontal well fracturing section is 7.83×10^4 m^3/d [98]. Shale gas with the predominant of CH_4 was also found in the Chengkou block of northeastern Chongqing and the Well TX1 of northern Guizhou area. The former has an average desorbed gas content of 1.07 m^3/t [99], while the latter has a gas content of 1.10 m^3/t-2.88 m^3/t with an initial daily yield of 0.3×10^4 m^3 [23]. In southeastern Chongqing, northwestern Hunan, and northern Guizhou areas (Upper Yangtze region), the Lower Cambrian shales not only have low gas content but also the gas compositions are mainly composed of N_2, with its percentage of 60%-90%. Figure 4 shows the gas compositions of representative shale gas wells with high content of N_2 in the Upper Yangtze region. The N_2 content has exceeded 60% and can be classified as the high-N_2 shale gas reservoirs [100]. At the same time, the $\delta^{15}N$ value of the Lower Cambrian shale gas in southern China is generally in the range of -2.6‰ to 0‰ [101–104]. The gas content of the Lower Cambrian shales in the Lower Yangtze area is also relatively low. For example, the highest GIP content of the Lower Cambrian shales from the Well XY1 in southern Anhui is 1.30 m^3/t, and the average GIP content is 0.94 m^3/t. The highest GIP content of the Lower Cambrian shales from the Well WY1 is only 0.15 m^3/t [24] (Table 2).

3. Pore Characteristics of the Lower Cambrian Shales

3.1. Pore Development Characteristics. The pores in shales are mainly divided into intergranular pores, intragranular pores, and OM pores [36, 37, 110–113]. Numerous studies have shown that the pores with diameter of >5-10 nm can be observed by scanning electron microscopy (SEM). The differences of pore development characteristics between the Lower Cambrian shales and the Lower Silurian shales in southern China are mainly reflected in the development degree of organic pores. Organic pores of the Lower Silurian shales are generally well developed, and the pores display the relatively larger diameters [84, 96, 114–116]. However, the development degree of organic pores in the Lower Cambrian shales is generally worse than that in the Lower Silurian shales. The organic pores in the Lower Cambrian shales are relatively small-sized and there display obvious differences in different regions [96, 117–120].

In the Jiaoshiba block of the Sichuan Basin, OM pores with clear morphological outlines, such as near-spherical, ellipsoidal, gneiss-shaped, pit-shaped, meniscus, and slit-shaped, are widely developed in the Lower Silurian shales. The pore diameter is mainly distributed between 2 nm and 1 μm, mostly of mesopores. The surface porosity of OM ranges from 10% to 50% with an average of 30% [84]. Abundant organic pores in organic-rich shales can form good gas conduction networks and improve the connectivity of shales.

Table 1: Geological and geochemical characteristics of the Lower Cambrian shales.

Regions	Sichuan Basin	Upper-Middle Yangtze Region (excluding Sichuan Basin)				Lower Yangtze Region
Distribution of organic-rich shales	Southern Sichuan, Southwestern Sichuan	Western Hubei	Chongqing	Northwestern Hunan	Northern Guizhou	Southern Anhui-Northern Jiangsu
Sedimentary face	Deep water shelf	Deep water shelf	Deep water shelf	Deep water shelf	Deep water shelf	Deep water shelf
Thickness of organic-rich shales (m)	60-300	20-80	20-200	50-120	40-100	20-100
Burial depth (m)	2500-6000	1000-5500	3500-5500	500-2500	1200-2400	4000-6000
EqVRo (%)	2.5-3.5	2.5-3	2.4-4.3	2-3.5	2.2-4.4	3.0-4.5
TOC (%)	2-3	2.4-3.2	0.4-9.9	0.3-6.4	1.7-9.6	2.3-14.5
Kerogen type	$I-II_1$	I	$I-II_1$	I	$I-II_1$	I
Reference	[12, 54, 57, 59, 63, 83–86]	[44, 45, 58]	[22, 51, 55, 87–90]	[21, 43, 52, 53, 65, 91]	[23, 56, 64, 92]	[24, 75, 93–95]

Table 2: Gas-bearing characteristics of the Lower Cambrian shales in the southern China (see Figure 2 for well locations).

Regions	Well	Formation	Gas content (m^3/t) Range/Average	Average content of CH_4 (%)	Average content of N_2 (%)	References
Sichuan Basin	W201	Qiongzhusi Fm.	1.10-3.51/2.01	95	nd	[96, 105, 106]
	JS1	Qiongzhusi Fm.	1.51-2.41/1.80	>90	<10	
	JY1	Qiongzhusi Fm.	1.02-4.68/2.03			
Western Hubei area	YY1	Shuijingtuo Fm.	0.58-5.48/2.05	90	8	[98]
	EYY1	Shuijingtuo Fm.	0.32-4.48/2.30	>90	<10	
	YD4	Shuijingtuo Fm.	0.50-3.13/1.54			
	ZD2	Shuijingtuo Fm.	0.23-4.45/2.15			
Chongqing area	YC1	Niutitang Fm.	0.03-1.12/0.22	16	84	[97]
	YY1	Niutitang Fm.	0.01-0.16/0.03	1	97	
	CY1	Shuijingtuo Fm.	0-1.23/0.65	>90	<10	[107]
	C1	Shuijingtuo Fm.	0.05-3.18/1.17	>90	<10	[55]
Northwestern Hunan area	CY1	Niutitang Fm.	0.03-2.10/1.02	9	72	[21, 79]
	CY-1	Niutitang Fm.	0.33-0.95	80	20	[97]
	HY1	Niutitang Fm.	0.10-0.29/0.02	13	84	
	BY2	Niutitang Fm.	0.11-0.71	9	91	[79]
Northern Guizhou area	CY1	Niutitang Fm.	0.30-1.80	>95	nd	[23, 92]
	TX1	Niutitang Fm.	1.10-2.88	80	16	
	YM1	Niutitang Fm.	0.10-0.40	nd	>90	
	HD1	Niutitang Fm.	0.09-1.31/0.42	<30	nd	
	HY1	Niutitang Fm.	0.32-2.00/1.50	>85	nd	[106, 108]
	FC1	Niutitang Fm.	0.40-3.50	84	5	[79]
Southern Anhui area	XY1	Hetang Fm.	0-1.30/0.94	<0.08	nd	[24, 109]
	WY1	Hetang Fm.	0-0.15	nd	nd	

Note: "nd" means not detected; CY-1 means Ciye1 Well, while CY1 means Changye1 Well in northwestern Hunan area.

OM pores are relatively developed in the Lower Cambrian shales from the Sichuan Basin, which generally occur within the infilling OM and residual kerogen. The diameter of organic pores in the Lower Cambrian shales is generally less than 50 nm, mostly of 10 nm to 30 nm, which is smaller than that in the Lower Silurian shales [96, 119]. The infilling OM is distributed in the intragranular pores of pyrite framboids and clay platelets, as well as the intergranular pores of quartz grains. Nanoscale pores are generally developed within the infilling OM [119].

The pore types of the Lower Cambrian shale reservoirs in western Hubei are mainly composed of organic pores, which usually have spongy, circular, or subcircular shapes. The pore diameter ranges from 4 nm to 84 nm, with an average of 12 nm [120]. The surface porosity generally varies from 5% to 20% [98]. Ma et al. [117] revealed that organic pores of the Lower Cambrian shales in the southeastern Chongqing were unevenly developed, and only some organic pores could be viewed. The organic pores generally have relatively small diameter, with mostly elongated or pinhole shapes, and the pore diameter varies from 10 nm to 50 nm. The organic pores are distributed in a dot network, and the connectivity is relatively poor. The surface porosity varies from 0.01% to 20%, with an average of 8%. The OM of the Lower Cambrian shales in northern Guizhou can be divided into two types, i.e., residual primary OM and secondary infilling OM [119]. The former displays a strip-shape or a large block, and organic pores can be rarely observed. However, the latter is mainly filled in the intragranular or intergranular pores of clay platelets and quartz grains, and spongy organic pores, with pore diameter of less than 50 nm, can be generally observed. Some of these organic pores are isolated, but some are interconnected. They are not uniformly developed within different parts of same OM particle, showing pore heterogeneity [118]. There have obvious differences in the development of OM pores in the Lower Cambrian shales.

3.2. Porosity and Pore Structure Characteristics. Significant differences in the porosity and pore structure are also occurred between the Lower Cambrian and Upper Ordovician-Lower Silurian shales in southern China. Previous works have showed that the Wufeng-Longmaxi Formation shales had the porosity of 1.46%-8.22%, with an average of 4.79%. The specific surface area varies from 6.2 m^2/g to 32.1 m^2/g, with an average of 17.56 m^2/g-23.84 m^2/g. Total pore volume varies from 0.02 cm^3/g to 0.07 cm^3/g, with an average of 0.041 cm^3/g. These porosity and pore structure parameters of the Lower Cambrian shales are significantly

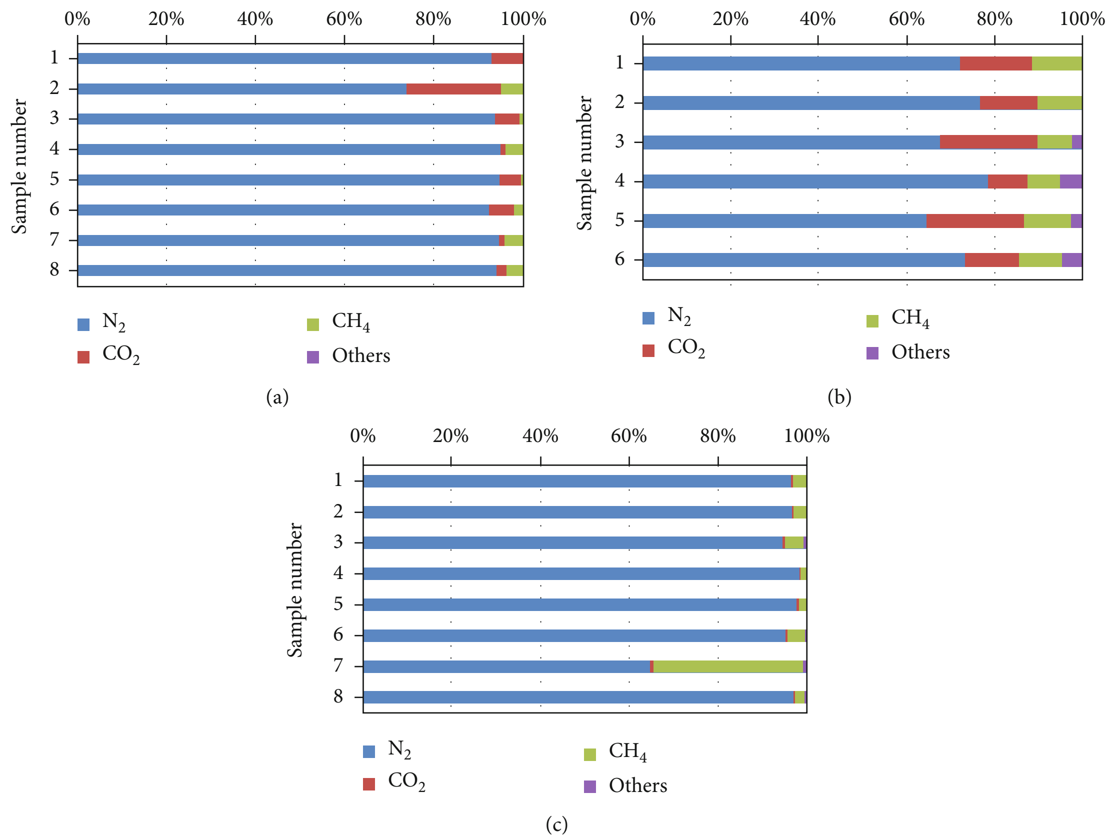

FIGURE 4: The gas compositions of the Lower Cambrian shale gas reservoirs in southern China. (a) Well YC9 in southeastern Chongqing, (b) Well CY1 in northwestern Hunan, and (c) Well TM1 in northern Guizhou (original data from Jiang et al. [79]; Wang et al. [92]; Jiao et al. [101], see Figure 2 for well locations).

lower than those of the Upper Ordovician-Lower Silurian shales. The porosity of the Lower Cambrian shales ranges from 0.3% to 3.33%, with an average of 1.62%. The specific surface area ranges from 0.8 m^2/g to 18.2 m^2/g, with an average of 7.06 m^2/g. Total pore volume ranges from 0.001 cm^3/g to 0.04 cm^3/g, with an average of 0.023 cm^3/g (Table 3).

The differences of pore structure between the Lower Cambrian and Upper Ordovician-Lower Silurian shales in different regions are also reflected in the pore diameter distribution. Wang et al. [121] had revealed that the micropore diameters of the Wufeng-Longmaxi Formation shales in the Sichuan Basin were mainly distributed around 0.35 nm, 0.46 nm-0.62 nm, and 0.83 nm and the nonmicropores were mainly composed of smaller mesopores. The pore diameter is generally distributed between 2 nm and 10 nm, while the macropores are rarely observed (Figure 5(a)). The pore structure of the Lower Cambrian Qiongzhusi shales in the Sichuan Basin is mainly composed of mesopores with a diameter of 2 nm-7 nm. The micropores also have a contribution to total pore volume, and the peaks are mainly distributed around 0.34 nm, 0.58 nm, and 0.83 nm. The macropores contribute little to total pore volume [122] (Figure 5(b)). The micropores of the Lower Cambrian shales in the northeastern Chongqing and northern Guizhou are well developed with pore diameter mainly ranging from 0.6 nm to 2 nm, while the nonmicropores are underdeveloped. However, there are some differences between these two regions. The nonmicropores in the northeastern Chongqing are mainly composed of small mesopores and the pore diameter is generally less than 10 nm, while the nonmicropores in the northern Guizhou are distributed between 2 nm and 100 nm [16, 123] (Figures 5(c) and 5(d)).

The contribution of organic pores to total pores in the Upper Ordovician-Lower Silurian shales is also significantly higher than that in the Lower Cambrian shales. Wang et al. [124] have found that organic porosity accounted for 13.9%-21.4% total micropore porosity in the Lower Silurian shales with an average of 17.61%, and organic porosity accounted for 10.4%-17.3% total micropore porosity in the Lower Cambrian shales with an average of 14.10%. Ma et al. [117] used the FIB-SEM 3D reconstruction method to compare the contribution of organic porosity in the Lower Silurian and Lower Cambrian shales in southeastern Chongqing and found that organic porosity of the Lower Silurian shales contributed 13.36%-23.51% total porosity. However, organic porosity of the Lower Cambrian shales only contributed 0.16%-5.93% total porosity. Total porosity of shales should be mainly composed of dissolved pores and intergranular pores of inorganic minerals. Nie et al. [125] suggested that the volume of organic pores of the Lower Silurian shales in southern Sichuan accounted for 30%-

Table 3: Pore structure parameters of the Lower Paleozoic shales in southern China (see Figure 2 for well locations).

Regions	Formation	Well	Buried depth (m)	Number of samples	Specific surface area (m^2/g) Range/Average	Total pore volume (mL/g) Range/Average	Porosity (%) Range/Average	References
Sichuan Basin	Wufeng-Longmaxi Fm.	N203	2170-2393	28	7.3-31.8/14.92	0.03-0.06/0.050	1.46-8.22/4.72	[96]
	Wufeng-Longmaxi Fm.	W201	1490-1552	6	6.2-20.2/14.00	0.03-0.07/0.047	3.79-6.75/5.96	
	Longmaxi Fm.	JY1	2288-2357	10	14.3-32.1/23.30	0.02-0.03/0.024	3.66-5.65/4.68	[126]
	Longmaxi Fm.	JY2	2552-2607	6	18.9-25.9/23.84	0.02-0.024/0.022	3.43-4.73/4.17	
	Qiongzhusi Fm.	W201	2705-2816	9	2.3-10.8/5.90	0.01-0.04/0.028	1.62-2.17/1.85	[96]
	Qiongzhusi Fm.	N206	1830-1887	9	4.5-9.6/7.64	0.03-0.04/0.038	0.47-2.57/1.52	
	Qiongzhusi Fm.	B1	2819-2977	9	3.9-7.0/5.14	0.02-0.03/0.028	1.50-1.87/1.64	
	Qiongzhusi Fm.	YS106	3260-3380	7	2.0-3.4/2.64	0.01-0.03/0.020	0.88-2.13/1.48	
Western Hubei	Shuijingtuo Fm.	YY1	1809-1871	6	7.2-18.2/14.00	0.01-0.04/0.025	0.96-3.33/2.08	[44]
Northwestern Hunan	Niutitang Fm.	CY1	795-1334	7	0.8-7.4/3.00	0.001-0.01/0.005	1.20-1.80/1.50	[21]
Northern Guizhou	Niutitang Fm.	TM1	1430-1457	4	10.6-15.2/12.80	0.008-0.01/0.010	nd	[127]
	Niutitang Fm.	CY1	1418-1440	4	8.3-13.3/11.43	0.004-0.02/0.008	nd	
Southern Anhui	Hetang Fm.	XY1	nd	15	4.3-7.5	0.006-0.012	0.30-2.82/1.47	[24]

Note: "nd" means not detected.

40% total pore volume, while the volume of organic pores of the Lower Cambrian shales in the northern Guizhou accounted for only 5%-40% total pore volume, with an average of about 20%.

4. Factors Controlling Porosity and Gas Content of the Lower Cambrian Shales

In general, there have two main factors affecting pore development and gas-bearing property of shales. One is the geochemical property of shales, including the TOC content, maturity, mineral composition, and kerogen type [113, 128–135]. Another is the geological characteristics of shales, including structural evolution and burial depth [75, 136–138]. The lower Cambrian shales in southern China are characterized by poor pore development, low CH_4 content, and high N_2 content, which may be related to the high maturity and strong tectonic deformation.

4.1. OM Maturity. The OM maturity can control gas compositions of shale gas reservoirs and affect their gas-bearing property [23, 47, 139–142]. The OM maturity can also affect pore characteristics of shales, especially the generation and evolution of organic pores, thus influencing reservoir property [130, 131, 143, 144]. The porosity of the Lower Cambrian shales in southern China is generally low (generally <2.0%). Different explanations are accounted for such low porosity values [22, 96, 145, 146], but high maturity is generally regarded to be a key factor [75, 120, 127, 147].

Borjigin et al. [148] believed that kerogen was a kind of high molecular polymer, which was composed of condensed cyclic aromatic nuclei linked with heteroatom bonds or aliphatic chains. During the evolution of kerogen, aliphatic chain bridges and heterocyclic functional groups are broken, releasing compound fragments of different sizes (e.g., bitumen with different relative molecular weights and volatile substances). When organic molecular fragments were released but not left completely from the kerogen parent, steric hindrance effect would be continued and condensation reaction would be hindered. The original storage space could be maintained within the disorderly arranged fragments. When fragments were discharged, the condensation reaction was strengthened, thus leading to reduction of pores due to the rearrangement and condensation of surrounding aromatic nuclei. Therefore, removal of aliphatic chains and heteroatom bonds can help to eliminate condensation barriers via hydrocarbon generation processes, such as decarboxylation and dealkylation [149]. Thus, the generation and evolution of organic pores are intimately related to thermal evolution process.

Previous studies have shown that organic porosity of shale could not increase monotonically with the increasing of OM maturity. Wang et al. [150] believed that organic porosity of shale generally increased with the increasing of OM maturity within the gas generation stage (Ro = 1.3% -2.0%), but organic porosity generally declined increased at the Ro > 2.0%. However, such a threshold (Ro = 2.0%) still remains controversial. More and more authors gradually believed that there were two peaks of hydrocarbon generation and the transition point of the second peak was generally 3.0% to 3.5% Ro [127, 142, 151, 152]. For example, Xu et al. [153] suggested that organic porosity increased rapidly with the increasing of OM maturity within the Ro < 3.0%, while it decreased significantly within the Ro ranging from 3.0% to 4.0%. The organic porosity slowly decreased at the Ro > 4.0% (Figure 6(a)). Consistent with organic porosity evolution of shale, pore structure of OM also undergoes a similar evolution trend. Thermal simulation experiments revealed that the evolution of organic pore structure could be divided into three stages, i.e., first stage (Ro ≈ 0.6%-2.0%), second stage (Ro ≈ 2.0%-3.5%), and third stage (Ro > 3.5%). During the first stage, the oil generated from kerogen would fill the intragranular and intergranular pores thus resulting in the reduction of pore spaces. Subsequently, the oil would be thermally cracked into gas and organic pores are formed. During the second stage, kerogen, pyrobitumen, and heavy hydrocarbon gas ($C_{2\text{-}5}$) would be further cracked into CH_4 and solid bitumen, thus resulting in a rapid increase of organic pores. During the third stage, with the further increase of temperature and pressure, the graphitization of OM would lead to the destruction of nanopore structure, and a large amount of nitrogen gas could be generated [139] (Figure 6(b)).

At the overmatured stage, original and secondary organic pores might be collapsed, shrunk, and compacted due to the escape of a large amount of hydrocarbon gas from organic pores, resulting in a large reduction of organic porosity [22, 47]. Other studies suggested that such a reduction might be related to the carbonization caused from strong condensation of aromatic structure of OM [82, 96, 145, 148, 154]. For example, Wang et al. [82] found that the porosity and resistivity of the Qiongzhusi and Longmaxi Formation shales in the Sichuan Basin displayed a sudden drop at the EqVRo > 3.5%, and they believed that the OM carbonization might destroy organic pore structure of shales. Wang et al. [155] also found that the laser Raman of OM in the Lower Cambrian shales showed obvious Raman peaks ($1347.2\,cm^{-1}$-$1606.4\,cm^{-1}$) pointing to carbonization when the EqVRo value was greater than or equal to 3.2%-3.5%.

The formation of high-N_2 shale gas in the Lower Cambrian shales in southern China is also closely related to maturity. Thermal simulation experiments have shown that shale samples can produce a certain amount of N_2 in the high-overmature stage [100, 156–160]. The N_2 can be formed from OM and inorganic minerals. The organic N_2 can be generated from sapropel and humic kerogen, while the inorganic N_2 can be derived from nitrogen-containing minerals, such as nitrate, nitrite, and ammonium [158, 161, 162]. The formation of inorganic N_2 is related to the ammoniation of ammonium-containing compounds via thermal catalysis [163], which is mainly occurred under the condition of the EqVRo < 3.4%. Thus, the N_2 generation potential in shales at this stage is mainly dependent on the content of inorganic nitrogen [164] (Figure 7). When a large amount of inorganic nitrogen is released, the hydrocarbon generation potential of kerogen

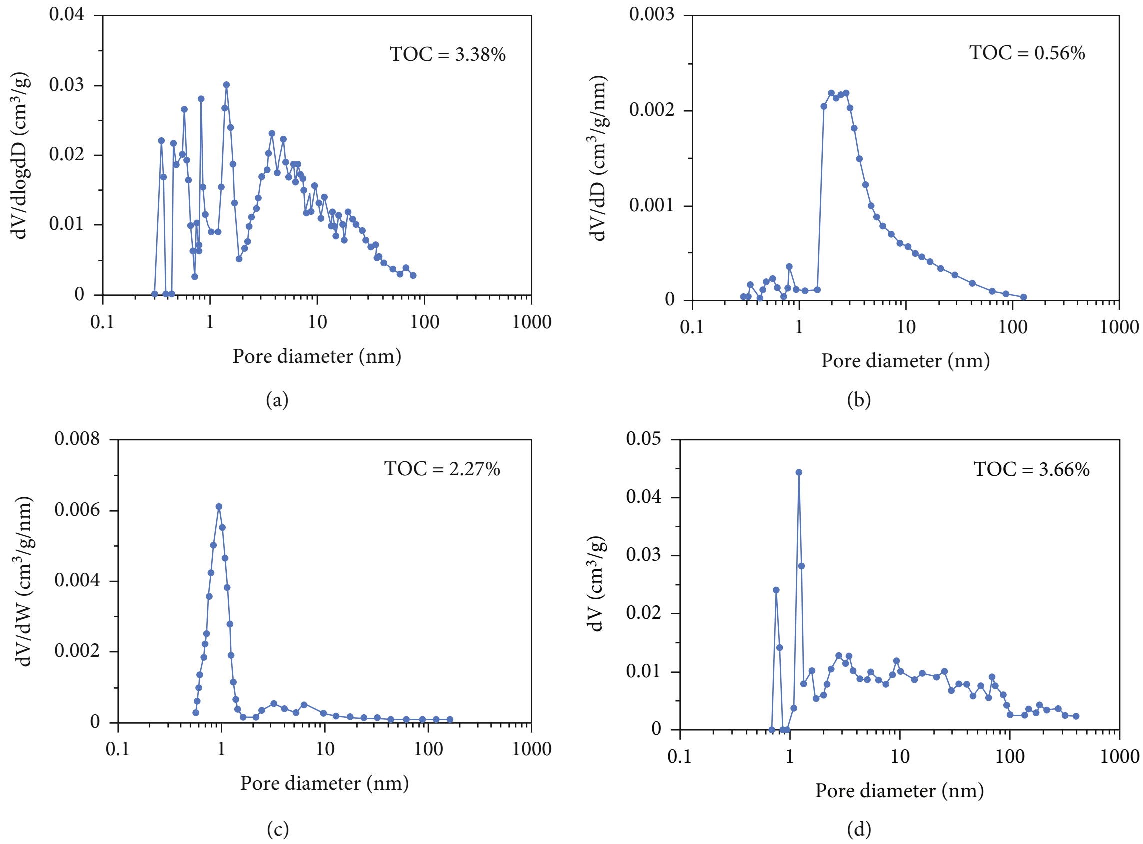

FIGURE 5: Pore size distribution of the Lower Paleozoic shales in southern China. (a) The Lower Silurian shale samples in southern Sichuan, (b) the Lower Cambrian shale samples in southern Sichuan, (c) the Lower Cambrian shale samples in northeastern Chongqing, and (d) the Lower Cambrian shale samples in northern Guizhou (modified from Wu et al. [16]; Wang et al. [121]; Yang et al. [122]; Zhu et al. [123]).

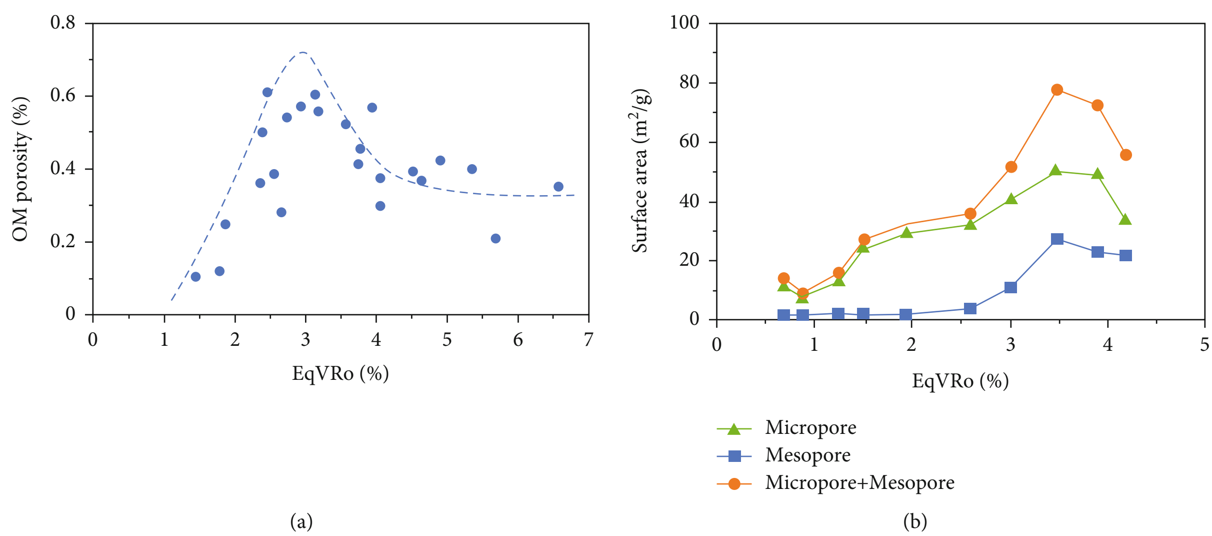

FIGURE 6: Cross-plots of the EqVRo value versus (a) OM porosity and (b) specific surface area (modified from Chen and Xiao [139]; Xu et al. [153]).

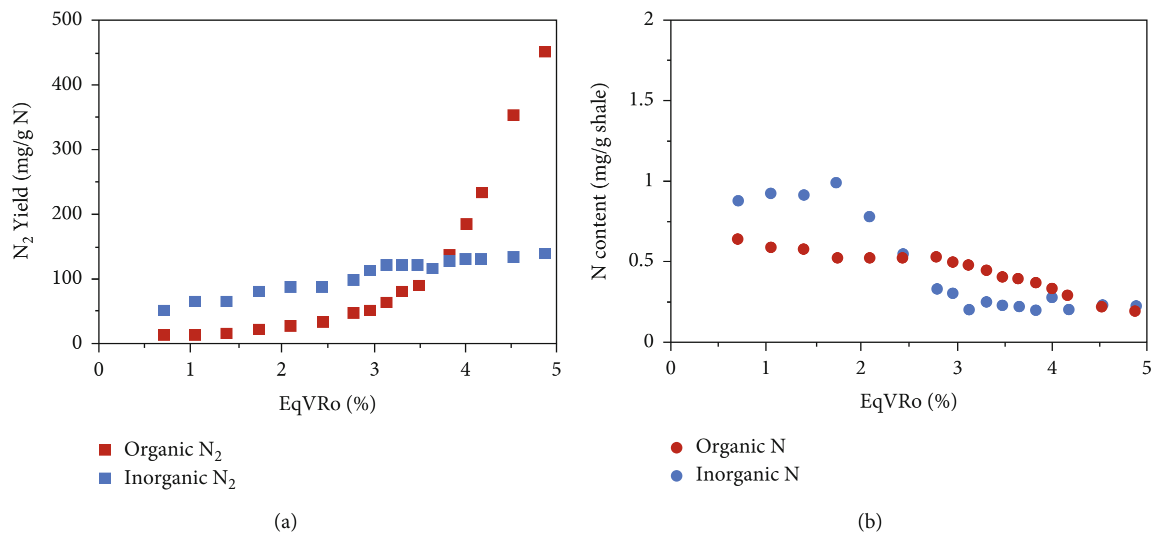

Figure 7: Cross-plots of the EqVRo value versus (a) N_2 yield and (b) molecular nitrogen yield during shale pyrolysis (modified from Gai et al. [164]).

is also exhausted and reactive nitrogen species will continue to be released.

Organic N_2 is mainly produced in the overmature stage of OM, and the high overmatured kerogen has the greater potential to generate high-N_2 shale gas [104, 159, 160, 164]. For example, Gai et al. [164] conducted pyrolysis analysis of the Lower Cambrian shales (EqVRo = 2.85%) from the southeastern Chongqing area and found that the N_2 and CH_4 production rates were increased rapidly at the EqVRo = 3.03%. When the EqVRo was greater than 3.4%, the yield of CH_4 began to decline and the yield of N_2 continued to increase. When the EqVRo was greater than 4.48%, the yield of N_2 exceeded that of CH_4 (Figure 8(a)). In geological conditions, the maturity of the Lower Cambrian shale gas with high-N_2 content had been proved to be higher. For example, the relative content of N_2 in shale gas reservoirs from the Well CY-1 (northwestern Hunan) and Well TM1 (northern Guizhou) is above 70% (Table 2), and the EqVRo value of these shales is basically greater than 3.0% [52, 104]. Wu et al. [103] revealed that the N_2 content of the Lower Cambrian shale gas reservoirs in the Well FC1 (northern Guizhou) was closely related to the degree of thermal evolution. When EqVRo was greater than 3.5%, the relative content of N_2 increases rapidly and was basically greater than 60% (Figure 8(b)).

The organic nitrogen content of shales is the material basis to influence the yield of N_2 at the high overmatured stage. Compared with the Lower Silurian shales, the Lower Cambrian shales in southern China usually have higher content of organic nitrogen. Liu et al. [165] showed that organic nitrogen content of the Lower Silurian shales in southern Sichuan ranged from 0.07% to 0.21%, with an average of 0.14%. However, organic nitrogen content of the Lower Cambrian shales in northern Guizhou varied from 0.23% to 0.60%, with an average of 0.45% [103]. As the degree of thermal evolution increases, organic nitrogen is gradually converted into N_2 [158]. In addition, the pores were preferentially filled with oil or solid bitumen at the high overmatured stage, so available pore spaces could be declined and gas flow could be restricted [143], resulting in the weak adsorption capacity of CH_4 on OM [166]. Under such a condition, the CH_4 can be easily escaped into shallow surface, while the adsorption capacity of N_2 on OM remains unchanged [167]. Therefore, the N_2 that generated at the late stage of hydrocarbon generation can be easily remained in the shale reservoirs and finally form the shale gas reservoirs with the characteristics of low content of hydrocarbon and high content of N_2. The combination of the abovementioned factors should be responsible for the formation of the high-N_2 shale gas reservoirs of the Lower Cambrian.

In a word, the OM of the Lower Cambrian shales in southern China is generally highly-matured, and the EqVRo value is generally >3.0% or even >3.5% in many areas [75]. The OM maturity may be mainly responsible for the low porosity of the Lower Cambrian shales, which exerts some influences in pore structure and gas-bearing property of shales in different blocks.

4.2. Tectonic Deformation. Compaction is also one of major factors affecting porosity of shales [35, 37, 125, 168, 169]. Previous studies have shown that intense compaction would reduce the porosity by 80%-90% for high overmatured shales [169, 170]. The Lower Cambrian shales in southern China not only underwent strong compaction but also experienced strong tectonic deformation, especially in the vast areas outside the Sichuan Basin [22, 75, 127]. The pores (especially organic pores) and pore structures would be changed due to the deformation of shales [171]. Previous studies have shown that porosity was positively correlated with brittle deformation and negatively correlated with ductile deformation [123]. Pan et al. [172] believed that brittle deformation of coal changes its chemical structure via the conversion of mechanical friction to thermal energy in the fault zone, while the ductile deformation mainly leaded to the deformation of the coal macromolecular structural unit and the dislocation creep of the aromatic ring via the

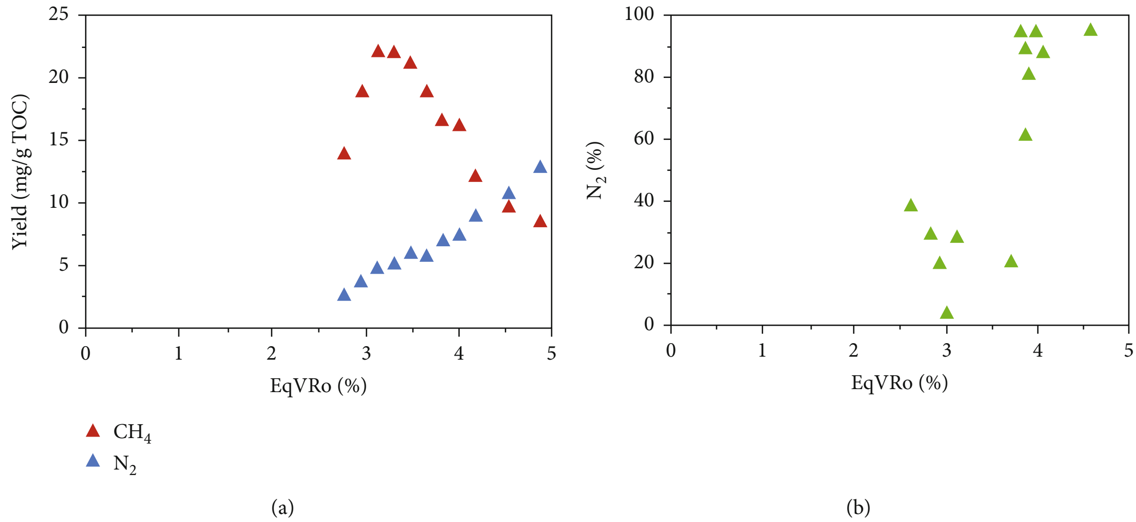

Figure 8: Cross-plots of the EqVRo value versus (a) CH_4, N_2 yield in Well YC2 and (b) N_2 percentage in Well FC1 (modified from Wu et al. [103]; Gai et al.[164]).

accumulation of strain energy, thus resulting in chemical structure destruction of the coal and the decrease of the porosity. Exploration practices showed that the deformation of shale under shallow burial (low pressure and low temperature) generally manifested as brittle deformation, while the shales gradually evolved from brittle-ductile transition zone to ductile zone under deep burial (burial depth of about 5000 m, high temperature and high pressure) [173]. Under such a condition, the ductility would be enhanced and organic pores would be seriously reduced due to compaction and extrusion [148].

TOC is an important factor affecting shale deformation [174], because OM has weaker resistant to compaction relative to mineral matrix. The compaction effect of shales with high TOC content is stronger than that of shales with low TOC content at the same conditions, then influencing the decrease ratio of the porosity [119]. Milliken et al. [37] found that the porosity of the Marcellus shales in North America increased with the increase of TOC content at the TOC < 5.6%, while the porosity will decrease at the TOC content exceeding 5.6% (Figure 9(a)). The Lower Cambrian shales in southern China also show a similar pattern, but the TOC threshold for the reversal of porosity is varied. For example, the TOC threshold for the Well XYA in southern Anhui, Well YD2 in western Hubei and Well HY1 in northern Guizhou appears to be about 2%, 3%, and 5%, respectively (Figure 9(b)). However, the porosity of the Lower Silurian shales in Well JYA and Well PYA continues to increase with the increase of TOC content when the TOC content ranges from 0.3% to 5.6% (Figure 9(b)). These results suggest that the relationship between TOC and porosity is complex and may be influenced by other factors, such as mineral composition, maturity, and tectonic deformation of shales. For example, Ma et al. [55] made a comparison of the porosity and pore structure between the Lower Cambrian deformed and nondeformed shales in northeastern Chongqing and found that the average porosity of deformed shales was 0.81% and the nondeformed shales was 1.24%. The average BET specific surface area and BJH pore volume of deformed shale are 7 m^2/g and 0.0073 cm^3/g, respectively, and those of nondeformed shale are 11.4 m^2/g and 0.011 cm^3/g, respectively.

The content of clay minerals of the Lower Cambrian shales in southern China is generally in the range of about 20% to 40%, and clay minerals mainly include illites, illite-smectite mixed layers, and a small amount of chlorites and kaolinites [22, 47, 118, 122, 176]. With the increasing burial depth, montmorillonites would be transformed into illites or illite-smectite layers. The specific surface areas of illites and illite-smectite layers are 7.1 m^2/g and 30.8 m^2/g, respectively, which are significantly lower than that of montmorillonites (76.4 m^2/g) [177]. Therefore, the specific surface area of shale mineral matrix would be significantly reduced during the transformation of clay minerals, although some pores and fractures are newly formed during such a transformation [178]. Clay minerals with high content will affect the mechanical property of shales and increase the plasticity of shales. The porosity and pore size of shales are more likely to be reduced without the support of rigid minerals and fluid pressure under high-pressure conditions [123, 148]. As shown in Figure 10(a), shale porosity is negatively correlated with clay mineral content. Brittle minerals have a significant positive impact on pore characteristics of shales [179].

Brittle minerals are generally stable and difficult to be dissolved, and their rigid frameworks can enhance the compaction resistance of shale. Thus, the pores (especially organic pores) could be effectively preserved under deep burial conditions [123, 127, 180]. The content of brittle minerals in the Lower Cambrian shales in southern China is high, and the quartz is the most common brittle mineral accounting for about 40%-60% of total minerals [56, 86, 120, 123]. As shown in Figure 10(b), the porosity is positively correlated with quartz content of shales. On the one hand, such a positive correlation may be resulted from widespread development of biological quartz fragments in marine shales [181–184]. The presence of biogenic silica

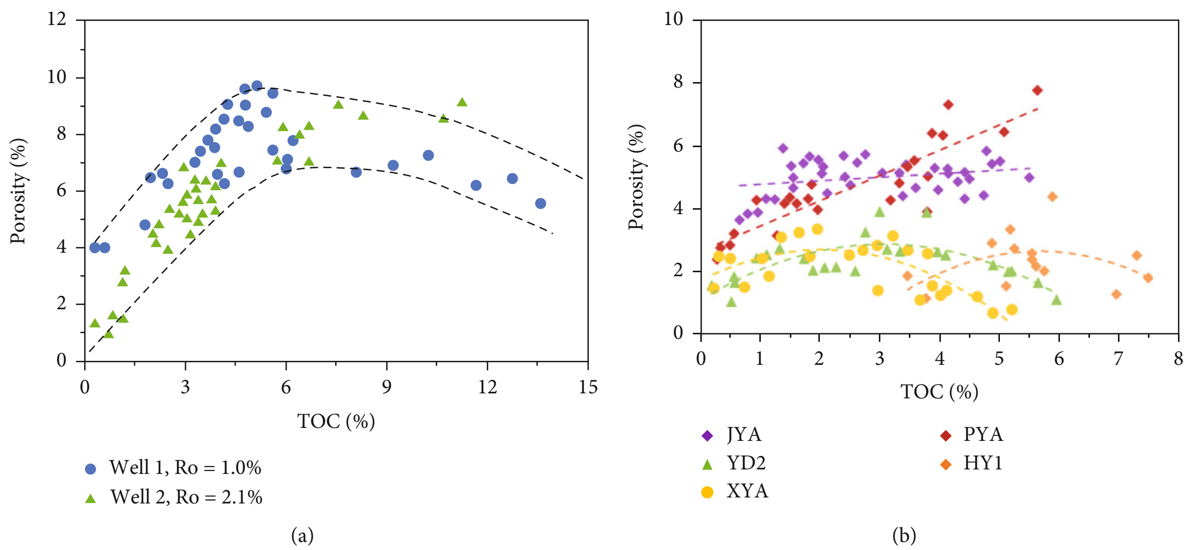

FIGURE 9: Cross-plots of the TOC value versus porosity of (a) the Marcellus shales in North America and (b) the Lower Paleozoic shales in South China. ((a) modified from Milliken et al.[37]; (b) data from Xiong, [119]; Xu et al., [153]; Chen et al., [175]).

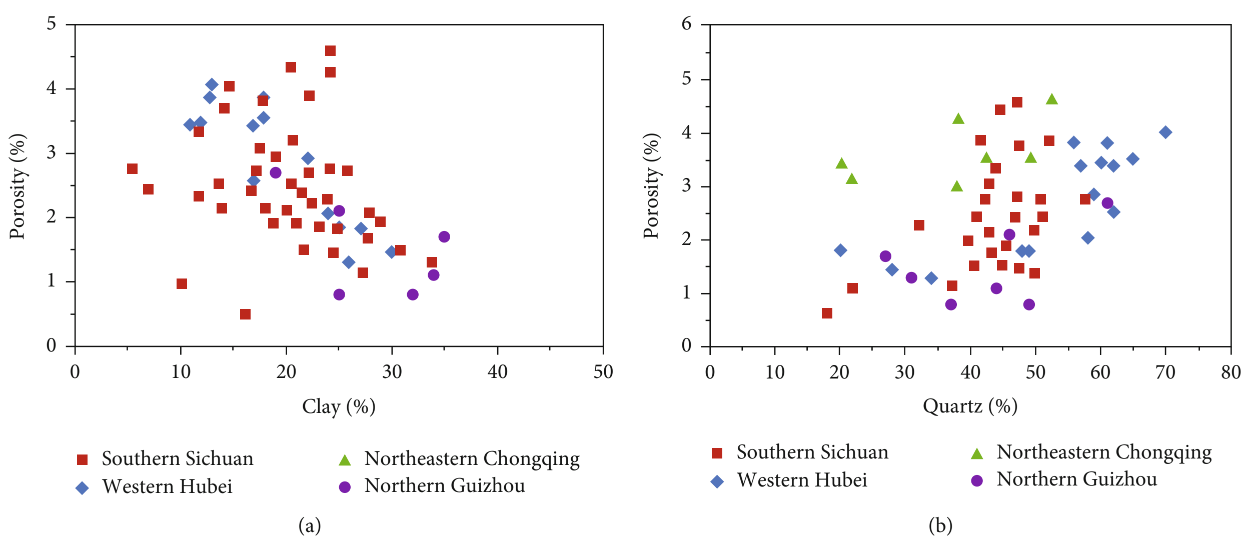

FIGURE 10: Cross-plots of the porosity versus (a) clay minerals and (b) quartz content of the Lower Cambrian shales in southern China (data from Li et al. [56]; Wang. [86]; Wei et al. [120]; Zhu et al. [123]).

can indirectly denote high TOC values [122], which can help increase porosity of shales, especially for OM pores. On the other hand, quartz has high hardness, which is beneficial to prevent pore collapse due to compaction. Some brittle grains are easily slipped or rotated at grain boundaries due to structural deformation, and they can move with respect to each other and form space in the weak zones of grains junction [185]. However, authigenic quartz can also plug pores in clay minerals which results in a certain reduction in porosity [122].

Except for few blocks with relatively stable structures in the Sichuan Basin, the Lower Cambrian shales in southern China have undergone strong tectonic deformation, which is considered to be one of major reasons accounting for the low content of natural gas or the enrichment of N_2 in shale gas reservoirs. For example, in the southeastern Chongqing area, on the one hand, relative sliding between hard ground (the underlying Dengying siliceous dolomites) and ductile organic-rich shales (the Lower Cambrian) can lead to the formation of detachment zones due to the compressive stress in the southeast direction [30, 167, 186, 187] (Figure 11); on the other hand, the Lower Cambrian shales were usually uplifted resulting in the formation of numerous thrust faults due to strong tectonic compression. The Lower Cambrian Niutitang shales were usually penetrated into the surface via the faults [167] (Figure 12). These faults and detachments together constitute a network for fluid intrusion and gas loss, resulting in the destruction of shale gas reservoirs, the loss of hydrocarbons, and the introduction of atmospheric N_2 into the shale reservoirs [100, 188]. Through the analysis of fluid inclusions, Jiao et al. [101] also found that the salinity of quartz and calcite inclusions in the

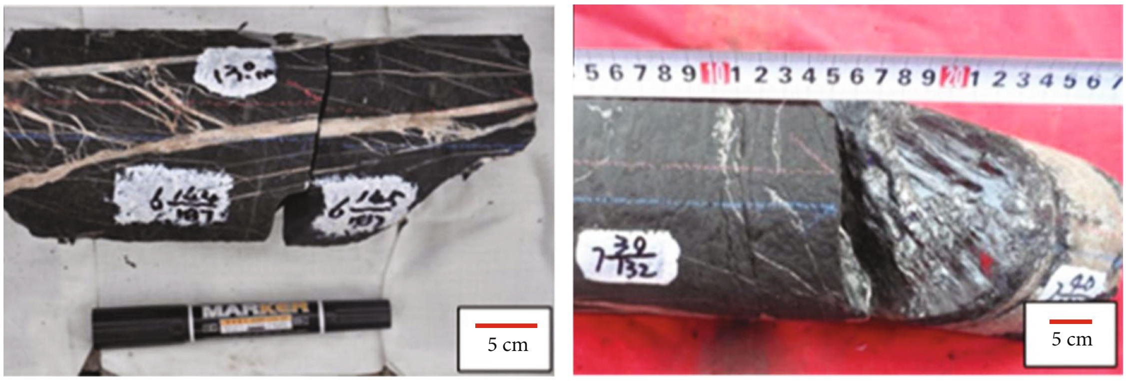

Figure 11: Detachment layers developed at the bottom of the Lower Cambrian in southeastern Chongqing area. (modified from Wang et al. [167]).

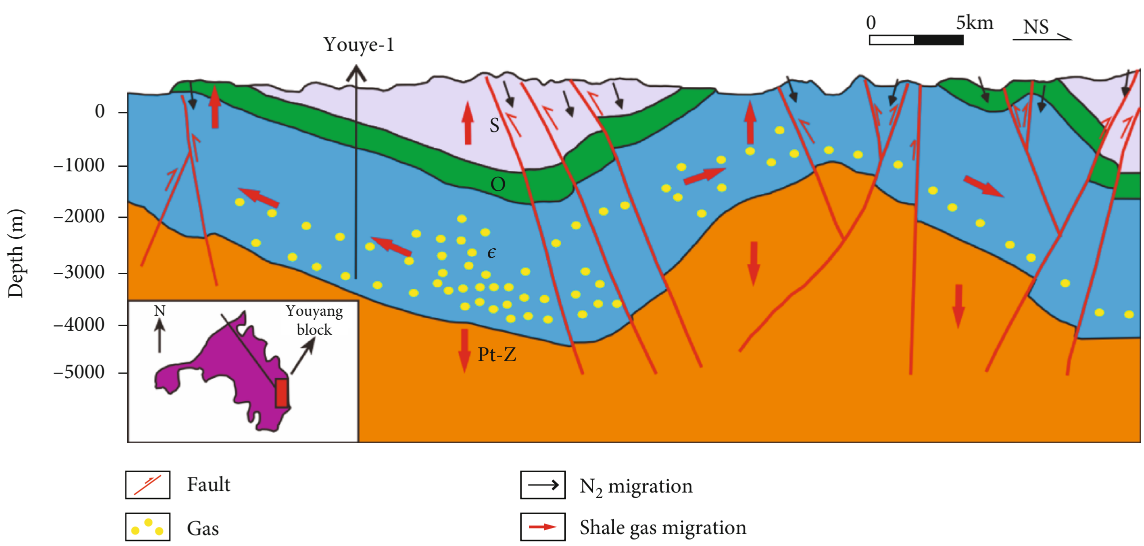

Figure 12: Geological section across Well Youye-1 in southeastern Chongqing area (modified from Wang et al. [167]).

Lower Cambrian shales from the southeastern Chongqing area varied greatly, ranging from 0.5% to 27% $NaCl_{ep}$, indicating that the fluid activity had been affected by atmospheric water precipitation [189, 190]. However, there have some exceptions. For example, the Lower Cambrian shales in the Chengkou block were strongly deformed and buried shallowly, but the gas content is relatively high (around $1\,m^3/t$) and the gas compositions are dominated by CH_4 (Table 2). Until now, there is no consensus on its formation mechanism. Zhu et al. [123] believed that the strong tectonic deformation led to the opening of intergranular pores, intragranular pores, microchannels, and microfractures in the Lower Cambrian shales, thus increasing the storage spaces. Han et al. [191] found that well-developed micropores, high pore-specific surface area, and strong gas adsorption capacity might be responsible for the enrichment of the Lower Cambrian shale gas in northeastern Chongqing. Meng et al. [192] thought that the Lower Cambrian shales are mainly characterized by micropores and small mesopores, with undeveloped mesopores, and their pores, such as OM-hosted pores and clay-hosted pores may be flattened by extrusion and/or compaction to have silt-like or layered shapes. This unique pore structure is obviously not conducive to gas loss and would play an important role in the preservation of shale gas. Ma et al. [22] suggested that three-dimensional connected pore system consisting of nanometer-sized intergranular pore spaces, aggregate pore spaces in clay flakes, and a pore network in the cleavage domains was developed in the Lower Cambrian shales, which might have a great contribution to preservation of shale gas in northeastern Chongqing.

4.3. Gas Occurrence and Pore Structure. Shale gas occurred in three forms: adsorbed, free, and dissolved phases, of which the adsorbed and free phases are predominated in shale gas [32]. Free gas mainly occurred in pores and natural fractures, while adsorbed gas mostly accumulates on the OM surface and micropores of inorganic minerals. Under the burial conditions, such two gas phases basically maintain a dynamic equilibrium of adsorption-desorption [193]. Previous works have reported that adsorbed gas content in typical shale gas plays (e.g., Lewis, Eagle ford, Marcellus, and Barnett) showed that the proportion of adsorbed gas varied greatly, ranging from 20% to 70% [194, 195]. The proportion

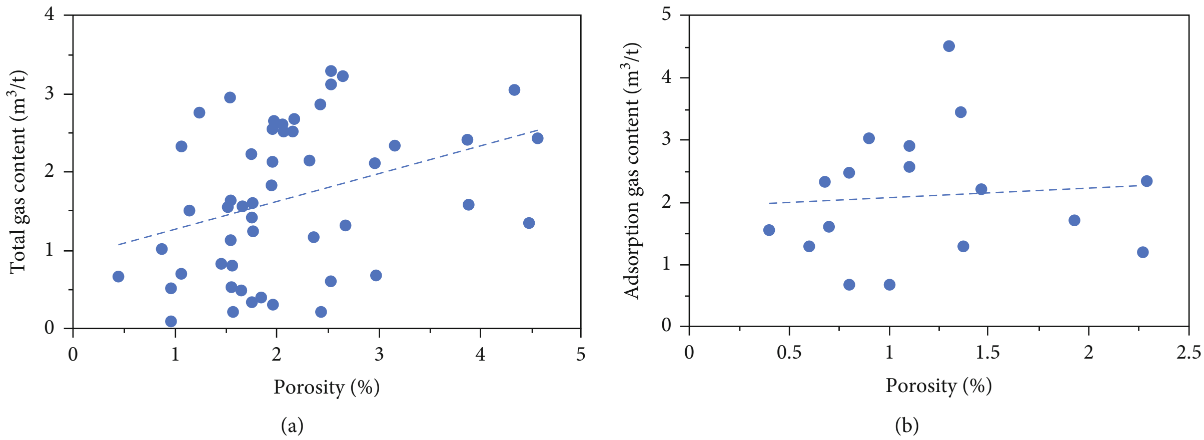

FIGURE 13: Cross-plots of porosity versus (a) total gas content of the Lower Cambrian shales in western Hubei and (b) adsorbed gas content of the Lower Cambrian shales in northeasernt Chongqing (data from Luo et al., [44]; Ma et al., [22]).

of adsorbed gas in the Lower Cambrian shale gas reservoirs in southern China is relatively high, ranging from 50% to 60%, while the proportion of free gas is relatively small, ranging from 40% to 50% [195–197].

The porosity has an important effect on gas content of shale reservoirs. In general, the higher the shale porosity is, the higher total gas and free gas content would be [3, 129, 198]. Luo et al. [44] showed a positive correlation between measured porosity and gas content of the Lower Cambrian shales in the western Hubei (Figure 13(a)). Wang et al. [150] found that the porosity of the Lower Cambrian shales (2.5%) was lower than that of the Lower Silurian (6%). At the burial depth of 3000 m, free gas content of the Lower Cambrian shales ($1.2\,m^3$/t-$2.3\,m^3$/t) is significantly lower than that of the Lower Silurian shales ($6.0\,m^3$/t-$6.8\,m^3$/t). They suggested that the porosity is the major factor controlling the differences of free gas content for these two successions of shales. However, the correlation between the adsorbed gas content and porosity is different from that of free gas. Chalmers et al. [124, 129] found no obvious correlation between the adsorbed gas content and porosity in organic-rich shales, which is consistent with the works of Ma et al. [22] (Figure 13(b)). The major reason is that CH_4 gas adsorption is mainly restricted by organic pores and organic pores of Lower Cambrian shales only contribute to a part of total shale pores.

Pore structure also has a certain influence on the storage capacity of shales, which in turn affects the gas content of shale [124, 199]. The specific surface area of pores greatly affects the storage capacity of shale and thus content of adsorb gas [200]. The specific surface area of shales is mainly provided by micropores and mesopores, but the specific surface area of macropores is far less than that of micropores and mesopores under the conditions of a united volume. Therefore, the more micropores and mesopores in the shale reservoirs are, the stronger adsorption capacity would be [201]. Previous works have revealed that the pore structure of the Lower Cambrian shales in southern China is mainly composed of micropores and mesopores, but macropores are relatively underdeveloped (Figure 5). Nanopores with the diameter less than 10 nm provide most of the specific surface area of the Lower Cambrian shales. There nanopores are mainly contributed from organic pores and control the adsorption capacity of shales [55, 122, 127, 195], leading to the predominate of adsorbed gas in total gas [22, 195–197].

5. Summary and Outlook

Geological and geochemical characteristics, gas-bearing characteristics, and their controlling factors of the Lower Cambrian shales in southern China have been extensively investigated. Several major conclusions can be drawn in the following:

(1) The Lower Cambrian organic-rich shales are mainly distributed in the Sichuan Basin, western Hubei, Chongqing, northwestern Hunan, northern Guizhou, and southern Anhui areas. The TOC content greatly varies, mostly of 2%-10%. The shales are highly and over matured, and the EqVRo value ranges from 2.5% to 6.0%. The gas content of shales also greatly varies, ranging from 0 to $5.48\,m^3$/t, and moreover, most shale gas reservoirs display low gas content or relative enrichment of N_2. Only few shale gas reservoirs in some blocks have high gas content and are enriched with CH_4.

(2) The porosity of the Lower Cambrian shales is very low, with an average of 1.47%-2.08%. Pore structure of shales is characterized by micropores and mesopores with smaller diameter (<10 nm). The porosity of shales is mainly contributed from mineral-related pores. The organic nanopores are relatively underdeveloped, which might have major contributions to micropore and mesopore with smaller diameter.

(3) Gas-bearing property of the Lower Cambrian shales might be controlled by geochemical characteristics, OM maturity, and tectonic deformation degree. The high content of N_2 in shale gas reservoirs might be mainly attributed to atmospheric source and/or

pyrolytic source, which may be linked to high maturity (EqVRo > 3.0%-3.5%) and or detachments and faults.

This review has provided some progresses of the exploration and development of the Lower Cambrian shale gas reservoirs in southern China, but it is still difficult to completely and objectively evaluate their resource potentials. In particular, major factors controlling pore development and preservation, as well as gas-bearing property of the Lower Cambrian shales remain unclear, which severely hinder the evaluation and exploration of shale gas in the southern China. With the increasing degree of shale gas exploration, some key research fields should be paid much more attentions, e.g., OM sources and types, OM formation and enrichment, generation-expulsion-evolution of hydrocarbon, the coupling relationship of mineral diagenesis, OM evolution and pore evolution, formation mechanism of nonhydrocarbon gases (e.g., N_2), competitive adsorption of nonhydrocarbon gases to CH_4, and its influences on gas content. In a word, the Lower Cambrian shale gas play with the low EqVRo value and the relatively stable tectonic conditions (especially deep-ultradeep burials) should be the favorable targets for the exploration and development of shale gas.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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