

Article

Study of the Methane Adsorption Characteristics in a Deep Coal Reservoir Using Adsorption Potential Theory

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Abstract: The gas adsorption characteristics in deep coal reservoirs are the focus of deep coalbed methane geology research. In order to reveal the adsorption characteristics in deep coal reservoirs and quantitatively characterize the amount of adsorbed methane in the deep coal seams, four coals were collected from the Permian Longtan Formation in southern Sichuan Province. Methane isothermal adsorption tests were carried out on the collected coal samples at 30 °C. The adsorption characteristic curve was established based on the data of the isothermal adsorption. The adsorption potential theory was used to predict the isothermal adsorption curves under different temperatures and the evolutionary relationship between the methane adsorption capacity and the coal seam burial depth in the C₁₇ and C₂₅ coal seams of the Permian in southern Sichuan Province, China. The results showed that the methane isothermal adsorption curve at 30 °C belonged to the Type I isotherm adsorption curve. The methane isothermal adsorption curves for various samples at 45 °C, 60 °C, and 75 °C were predicted based on the uniqueness of the methane adsorption characteristic curve. The amount of adsorbed gas in deep coal reservoirs was comprehensively controlled by pressure and temperature. The pressure showed a positive effect on the amount of methane adsorbed, while the temperature showed a negative effect on the adsorption of methane. The negative effect of temperature became more significant with the increase in pressure. The results of the study are beneficial for further promoting the exploration and development of deep coalbed methane in the southern Sichuan Province of China.

Keywords: adsorption potential; adsorption characteristics; deep coalbed methane; southern Sichuan Province



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1. Introduction

Currently, energy demand is constantly increasing due to the increase in population. To meet the growing energy demand, the extraction of coalbed methane has become necessary. Globally, the total amount of coalbed methane in coal (1730 T tons) is approximately 30,000 TCF [1]. As is well known, coalbed methane is present in the adsorbed state within the pores of a coal matrix and in a free state in coal pores and fractures, along with a small amount in a dissolved state in coal seam water [1–3]. The accumulation of coalbed methane is a prerequisite and foundation for its exploration and development [4]. From the 1980s to the early 21st century, the exploration of coalbed methane with depths ranging from 1500 to 3000 m was conducted in North America [5–7]. Scientists estimated the resources of coalbed methane in the major basins of North America and pointed out that the development of deep coalbed methane was an important direction for developing unconventional natural gas in the future [8]. The test project consisting of 65 coalbed methane wells (with coal seams buried at depths of 1635–2591 m) achieved success in the Piceance Basin [7,9], indicating the prospects for the exploration and development of deep coalbed methane. In China, a large-scale deep coalbed methane block was built on the

southern margin of Yanchuan [10]. Since 2021, several coalbed gas wells with a depth of more than 2000 m have produced tens of thousands of cubic meters of gas per day in the Junggar Basin and the eastern margin of the Ordos Basin [11–13]. Therefore, the study of methane adsorption characteristics in deep coal reservoirs is a hot topic, which can help predict the total exploitable methane content in deep coal reservoirs [14,15].

Experimental testing and numerical simulation are the main approaches used to study methane adsorption on coal. Previous studies have shown that the methane adsorption capacity on coal is controlled by internal factors such as coal properties, macerals, rank, deformation, and moisture [16–21] and various other external factors such as temperature and pressure [22–24]. Yee et al. [16] found that the adsorption capacity of dry coal for methane varied during various stages of the rank of coal. For example, in the low-rank stage of coal, the methane adsorption capacity on coal increased with the increase in the rank of coal. In the middle- to high-rank stage of coal, the methane adsorption capacity on coal showed a U-shaped variation trend with the increase in the coal rank. There is a minimum adsorption capacity near the highly volatile bituminous coal. Vitrinite has the strongest methane adsorption capacity, while exinite has the weakest methane adsorption capacity among the three petrography compositions [17–19]. The micropores (<2 nm) often result in the largest proportion of the total specific surface area and are the major contributors to the adsorption of methane [17,20]. Coal with a high content of aromatic carbon has a strong adsorption capacity, whereas coal with aliphatic structures and oxygen-containing functional groups exhibits a weaker capability for methane adsorption [25,26]. Pressure has a positive effect, whereas temperature has a negative effect on the adsorption of methane [13]. There are various models used to describe the adsorption of methane on coal, including the Langmuir model, BET model, D-A model, and D-R model [1,3,4,27,28]. The Langmuir model has been widely applied due to its simplicity and the practical value of its parameters.

With the continuous deepening of coalbed methane exploration and development, the adsorption characteristics and reservoir formation mechanism of deep coalbed methane have become increasingly valued. However, changes in pressure and temperature with the increase in burial depths affect the adsorption characteristics of coal and the occurrence state of coalbed methane. Physical simulation experiments can help researchers understand the adsorption situation under specific temperature and pressure conditions. However, it is difficult to simulate the gas adsorption characteristics of coal under high-temperature conditions. Therefore, it is difficult to describe the temperature and pressure conditions of methane adsorption characteristics on coal in deep reservoirs. The adsorption potential theory of non-polar carbon materials for gas adsorption fully characterizes the influence of temperature and pressure on the methane adsorption capacity of coal. In recent years, it has been applied to the evaluation theory of methane adsorption capacity on coal and in reservoirs. Therefore, based on the experimental data of methane isothermal adsorption on coals collected from southern Sichuan Province, China, the methane adsorption characteristic curves were constructed to predict the methane adsorption characteristics on coal under different temperature conditions. Moreover, the volume of the adsorbed methane in deep coal reservoirs was also predicted.

2. Adsorption Potential Theory

2.1. Theoretical Basis

The early adsorption potential theory mainly includes the following three aspects [29–31]. Firstly, there is a gravitational field within a certain space on the surface of an adsorbent just as there is a gravitational field on Earth that envelops air into an atmosphere. Gas molecules will be adsorbed once they fall within this gravitational field. The space in which the gravitational field operates is called the adsorption space. The density of the adsorbed gas in the adsorption space decreases with the increase in the distance from the surface. The density of the adsorbed gas at the outermost edge of the adsorption space is no longer different from that of the external gas. The maximum space in which the gravitational field acts is called the ultimate

adsorption space. Secondly, there is adsorption potential everywhere in the adsorption space. The adsorption potential is defined as the work required to adsorb 1 mol of gas from an infinite distance to a certain point. Thirdly, the adsorption potential is independent of the temperature, that is to say, the relationship between the adsorption potential and the adsorption space remains unchanged under any temperature conditions. Therefore, the relationship curve is called the adsorption characteristic curve.

2.2. Adsorption Potential

The adsorption potential theory is a thermodynamic theory proposed by Polanyi and is applicable to physical adsorption. The adsorption potential theory suggests that the interaction force between the gas and the solid is a dispersion force and that the distribution curve of the adsorption potential according to the adsorption space is unique and called the adsorption characteristic curve. This theory has been successfully applied to the adsorption of gases onto non-polar carbonaceous adsorbents [29–31]. As a special porous carbonaceous material, coal follows the Langmuir monolayer theory for gas adsorption, which has been confirmed by a large number of adsorption tests.

The relationship between the adsorption potential and the pressure is established by the adsorption potential theory and is given by Equation (1):

$$\varepsilon = \int_{P_i}^{P_0} \frac{RT}{P} dP = RT \ln \frac{P_0}{P_i}, \quad (1)$$

where P is the equilibrium pressure (Mpa), ε is the adsorption potential (J/mol), P_0 is the virtual saturated vapor pressure of methane (Mpa), P_i is the equilibrium pressure of ideal gas at constant temperature (Mpa), R is the universal gas constant, with a value of 8.3144 J/(mol·K), and T is the measured temperature (K).

Due to the adsorption of methane on the surface of coal being above the critical temperature, the saturated vapor pressure under critical conditions loses its physical significance. This article adopts the empirical correlation for virtual saturated steam pressure under supercritical conditions [30], as given by Equation (2):

$$P_0 = P_c \left(\frac{T}{T_c} \right)^2, \quad (2)$$

where P_c is the critical pressure of methane, with a value of 4.62 Mpa, and T_c is the critical temperature of methane, with a value of 190.6 K.

2.3. Adsorption Space

The adsorption space refers to the place in coal where methane can be adsorbed. The adsorption space is calculated based on isothermal adsorption data using Equation (3):

$$w = V_{ad} \frac{M}{\rho_{ad}}, \quad (3)$$

where w is the adsorption space (cm³/g), V_{ad} is the measured adsorption capacity (mol/g), M is the methane molecular weight (g/mol), and ρ_{ad} is the methane adsorption phase density (g/cm³), which can be calculated by Equation (4) [32]:

$$\rho_{ad} = \rho_b \exp[-0.0025 \times (T - T_b)], \quad (4)$$

where ρ_b is the liquid density of methane at normal boiling point, with a value of 0.466 g/cm³, T_b is the boiling point temperature of methane, with a value of 111.6 K, and T is the measured temperature (303.2 K).

2.4. Adsorption Characteristic Curve

The adsorption characteristic curve can be obtained based on the adsorption potential and its corresponding adsorption space. The relationship between the adsorption potential and the adsorption space can be described using Equation (5):

$$\varepsilon = a + bw + cw^2 + dw^3, \quad (5)$$

where a , b , c , and d are constants.

3. Experimental

3.1. Sample Preparation

The coal samples were collected from the XW Coal Mine, WH Coal Mine, and ZK drilling of southern Sichuan Province, China (Figure 1). The oldest exposed strata in the research area are the Sinian system. Except for the sporadic distribution of the Neogene Pleistocene series, the latest strata are the Cretaceous system, with the absence of the Cambrian Silurian and Carboniferous systems. The main strata distributed in the area are the Triassic Jialingjiang Formation and Feixianguan Formation, as well as the Permian Changxing Formation, Longtan Formation, and Maokou Formation.

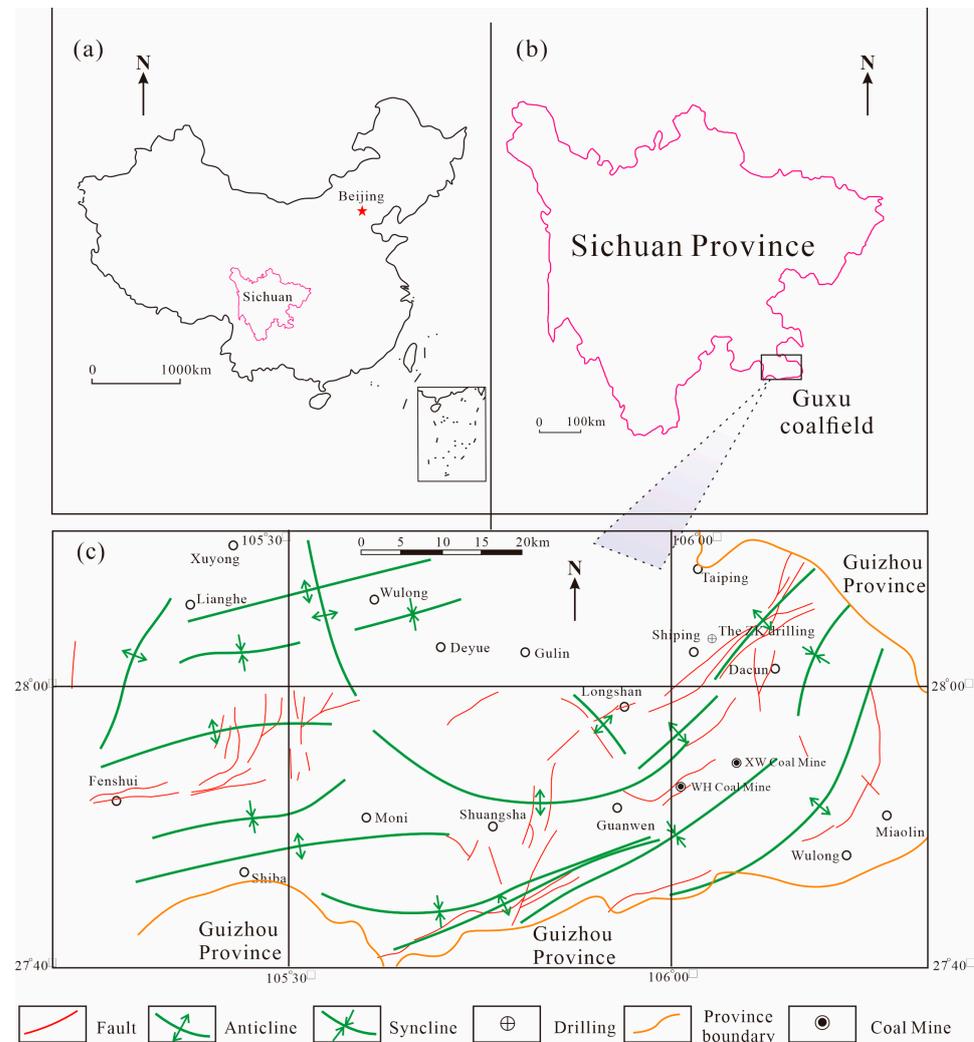


Figure 1. Map and the location of coal samples collected in the current work: (a) map of China, (b) map of Sichuan province, (c) structure outline map and the location of coal samples collected.

Among them, the coal-bearing strata of the Upper Permian are mainly the Longtan Formation, which is a set of marine continental sedimentary systems that are mainly composed of terrestrial facies. The thickness of the strata is about 76.25–130.11 m, with an average value of 92.33 m. It is composed of clastic rocks, mudstones, and coal seams (Figure 2). The top formation contains a small amount of bioclastic limestone or mudstone, while the upper and lower parts contain pyrite nodules. There are a total of 11–24 coal seams, with nine coal seams that can be compared, C₁₁, C₁₃, C₁₄, C₁₅, C₁₇, C₂₀, C₂₃, C₂₄, and C₂₅. Among them, there are two stable and minable coal seams (C₁₇ and C₂₅) in the entire area, whereas six coal seams (C₁₃, C₁₄, C₁₅, C₂₀, C₂₃, and C₂₄) are mostly and locally minable. The total thickness of the coal seams is 10.16 m, with a coal content coefficient of 12.37% and a minable coal content coefficient of 9.95%. The gas content is 4.76–39.38 m³/t, with an average of 14.87 m³/t.

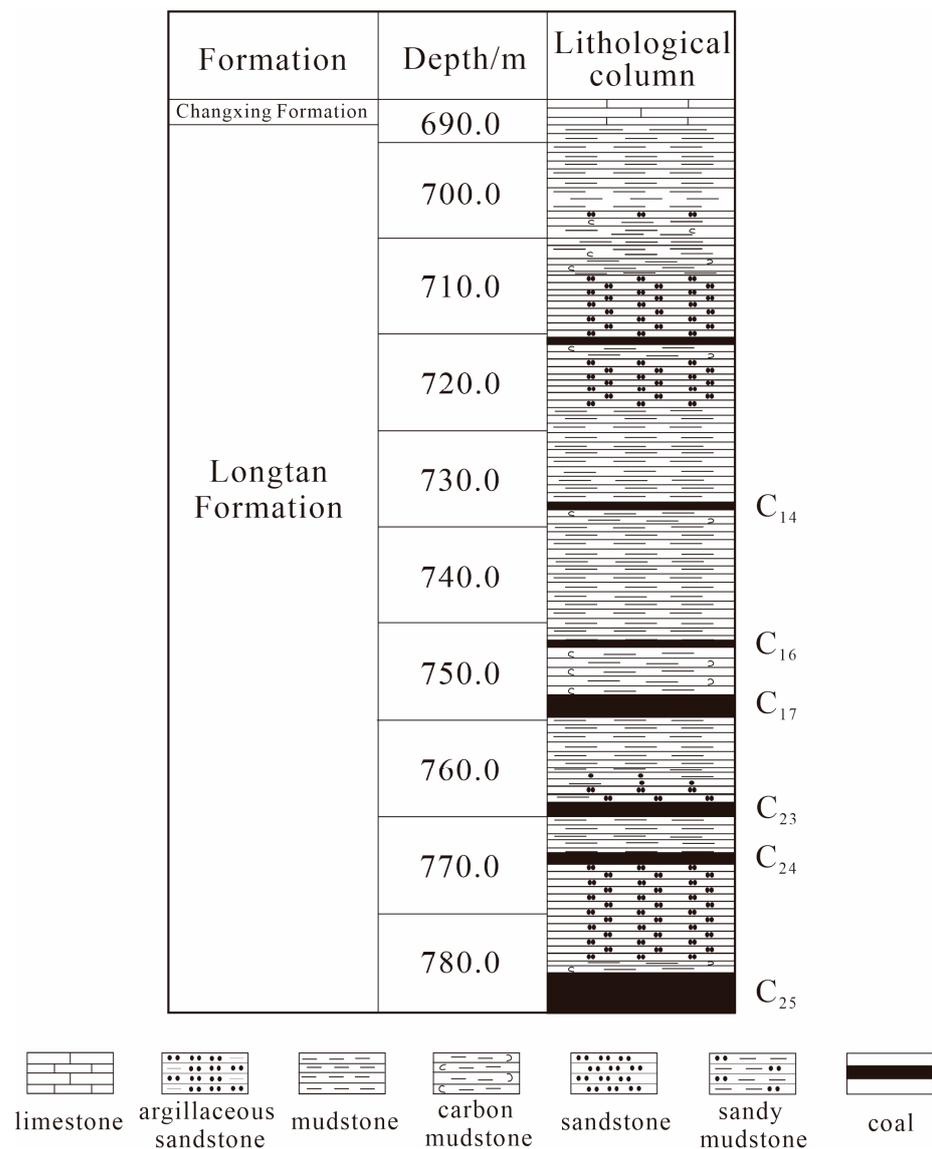


Figure 2. Stratigraphic column of the ZK drilling in southern Sichuan (China).

After being collected and before being transported to the laboratory for further analysis, all of the coal samples were wrapped in black polyethylene bags. The physical properties of the samples used in the current study are listed in Table 1. The samples are anthracites. Moreover, the value of M_{ad} lay within the range of 1.16–2.06%, whereas that of A_{ad} lay

within the range of 12.21–33.16%. Furthermore, the value of V_{ad} lay within the range of 8.23–10.32%.

Table 1. Information on the coal samples used in the current work.

Samples	Coal Seam	Coal Type	M_{ad} (%)	A_{ad} (%)	V_{ad} (%)
XW1	C ₁₇	anthracite	2.06	33.16	9.72
XW2	C ₂₅	anthracite	1.16	14.46	10.32
WH	C ₁₇	anthracite	2.35	12.21	8.94
ZK	C ₂₅	anthracite	1.83	16.79	8.23

Note: M_{ad} —air-dried moisture, A_{ad} —air-dried ash, V_{ad} —air-dried volatile.

3.2. Isothermal Adsorption Test

Methane isothermal adsorption tests were conducted on the samples using a laboratory-scale commercial isotherm system on an H-sorb2600 adsorption system. The samples were crushed to 60–80 mesh in accordance with the Chinese Standard GB/T19560-2008, and then they were dried at 110 °C until the weight loss was less than 1%. After drying, the samples were ready for the isothermal adsorption tests. The measurements were carried out at 30 °C, and each isothermal adsorption curve included 10 different pressures. Adsorption equilibrium at each pressure point was maintained for 1 h during the test. The adsorbate was 99.99% methane. Approximately 10 g of each sample was tested.

4. Results and Discussion

4.1. Methane Adsorption Characteristic

As shown in Figure 3, the methane adsorption curves had the same characteristics. The volume of methane adsorbed on the coal increased rapidly with the increase in pressure until 3 Mpa. The rate of increase in adsorbed methane gradually slowed down when the pressure lay within the range of 3–5 Mpa. The rate of increase in adsorbed methane was not significant when the pressure exceeded the value of 5 Mpa, and therefore, it was inferred that the system had reached its saturated state at this stage.

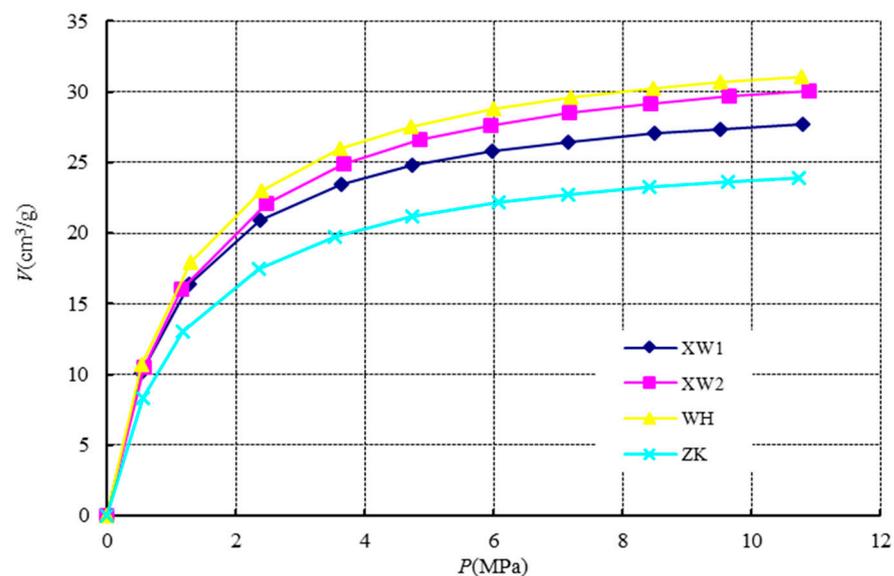


Figure 3. Measured methane isotherms at 30 °C.

Based on the curve characteristics of the methane adsorption on samples, the Langmuir model was used to describe the isothermal adsorption curves of the samples, as given by Equation (6):

$$V = \frac{V_L P}{P_L + P} \quad (6)$$

where V is the methane adsorption volume at equilibrium (cm^3/g), P is the equilibrium pressure (Mpa), V_L is the Langmuir volume under the standard state with a pressure of 101 kPa and a temperature of 273.15 K, and P_L is the Langmuir pressure (Mpa).

The Langmuir constants of the samples are presented in Table 2. In the present study, the Langmuir volume lay within the range of 26.67–34.56 cm^3/g , whereas the Langmuir pressure lay within the range of 1.10–1.28 Mpa. Fitting had a high correlation coefficient.

Table 2. The Langmuir constants of the samples.

Sample	V_L (cm^3)	P_L (MPa)	R^2
XW1	30.54	1.10	0.9573
XW2	33.59	1.28	0.9825
WH	34.56	1.20	0.9782
ZK	26.67	1.23	0.9659

Note: Dry basis.

4.2. Adsorption Characteristic Curve

In order to obtain the methane adsorption characteristic curves, the volumes of the adsorbed methane on coals under different pressures at the temperature of 30 °C were calculated according to the adsorption constants of the Langmuir model. Then, the adsorption potentials and adsorption spaces under different pressures at 30 °C were calculated. Following this, the adsorption characteristic curves were obtained (Figure 4). The fitting equations were obtained based on the adsorption characteristic curves as given by Equations (7)–(10). It can be seen that the univariate cubic equation could satisfactorily fit the relationship between the adsorption potential and the adsorption space with an R^2 of 0.9998.

$$\epsilon_{XW1} = 1.43 \times 10^4 - 4.22 \times 10^5 w + 7.88 \times 10^6 w^2 - 6.90 \times 10^7 w^3 \quad R^2 = 0.9998 \quad (7)$$

$$\epsilon_{XW2} = 1.41 \times 10^4 - 3.92 \times 10^5 w + 6.64 \times 10^6 w^2 - 5.22 \times 10^7 w^3 \quad R^2 = 0.9998 \quad (8)$$

$$\epsilon_{WH} = 1.42 \times 10^4 - 3.77 \times 10^5 w + 6.21 \times 10^6 w^2 - 4.77 \times 10^7 w^3 \quad R^2 = 0.9998 \quad (9)$$

$$\epsilon_{ZK} = 1.42 \times 10^4 - 4.9 \times 10^5 w + 1.05 \times 10^7 w^2 - 1.04 \times 10^8 w^3 \quad R^2 = 0.9998 \quad (10)$$

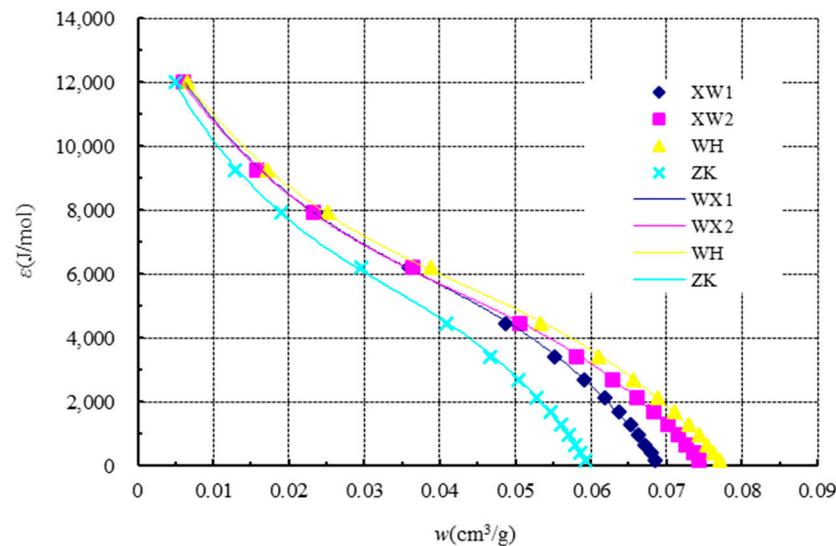


Figure 4. Predicted adsorption characteristic curve of methane on coal samples using adsorption potential theory.

In the present study, the maximum adsorption potential of around 12,000 J/mol and the maximum adsorption space of 0.059–0.008 cm^3/g (Figure 3) were obtained. There was

a significant difference in the adsorption characteristic curves among coals from coal mines and the drilling well. Compared to drilled coal samples, the coal samples collected from coal mines had larger adsorption spaces at the same adsorption potential. This phenomenon may be due to the significant differences in the sample collection and processing process.

4.3. Application of the Adsorption Characteristic Curve

A previous study [33] has shown that the methane adsorption characteristic curve for a specific coal sample is unique. The absolute adsorption capacity at any temperature and pressure can be calculated based on the equation of the methane adsorption potential characteristic curve. In the present study, the methane isotherm adsorption curves of the four coals at 45, 60, and 75 °C were predicted (Figure 5).

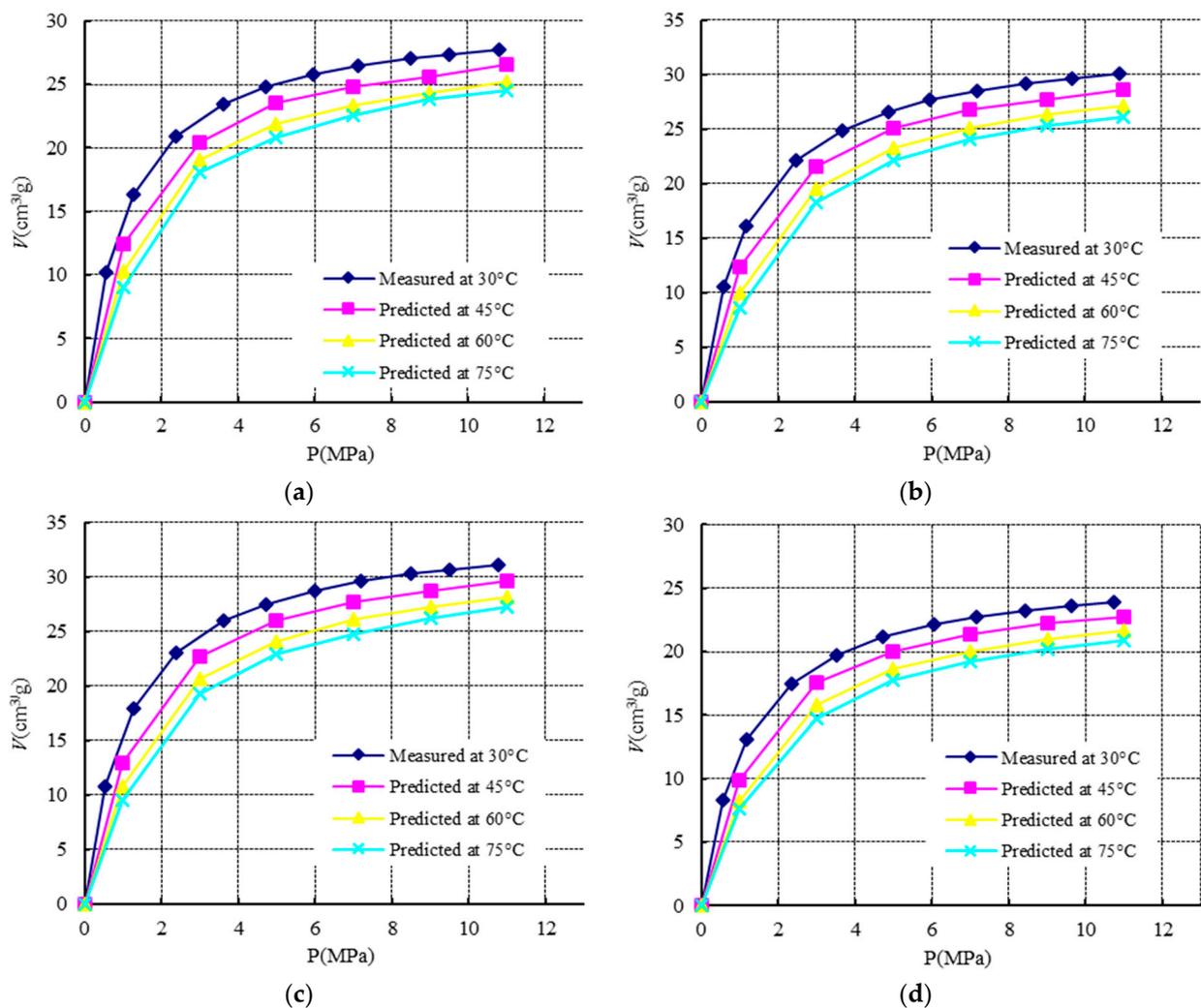


Figure 5. Measured and predicted methane isotherms at different temperatures: (a) XW1; (b) XW2; (c) WH; (d) ZK.

As shown in Figure 5, the adsorption capacity of methane on coal continuously weakened with the increase in temperature, which is completely consistent with previous studies. Meanwhile, it was found that predicting the adsorption isotherm curve at high temperatures still followed the Langmuir model. Then, the Langmuir constants were calculated using the Langmuir model based on the predicted results of the amounts of methane adsorbed under certain pressure values (Table 3).

Table 3. The prediction results of the methane isothermal adsorption on the samples at different temperatures.

Sample	T (°C)	V_L (cm ³ /g)	P_L (MPa)	R ²
XW1	45	29.79	1.38	0.9999
	60	29.28	1.74	0.9989
	75	29.20	2.05	0.9985
XW2	45	32.88	1.61	0.9998
	60	32.66	2.12	0.9990
	75	32.53	2.50	0.9980
WH	45	33.89	1.55	0.9996
	60	33.42	1.96	0.9992
	75	32.94	2.25	0.9988
ZK	45	26.21	1.58	0.9993
	60	25.80	2.00	0.9992
	75	25.24	2.20	0.9985

Note: Dry basis.

Meanwhile, the data on ground temperature and pressure at different burial depths were obtained based on the measured geological parameters of the ground temperature gradient and pressure gradient in the southern Sichuan region. In this study, the geothermal gradient of 2.45 °C/100 m and the pressure gradient of 0.98 MPa/100 m were used. The volumes of methane adsorbed on coals were obtained as mentioned earlier. Finally, the relationship between the volume of methane adsorbed on coal and the burial depth of the coal seam was obtained (Figure 6).

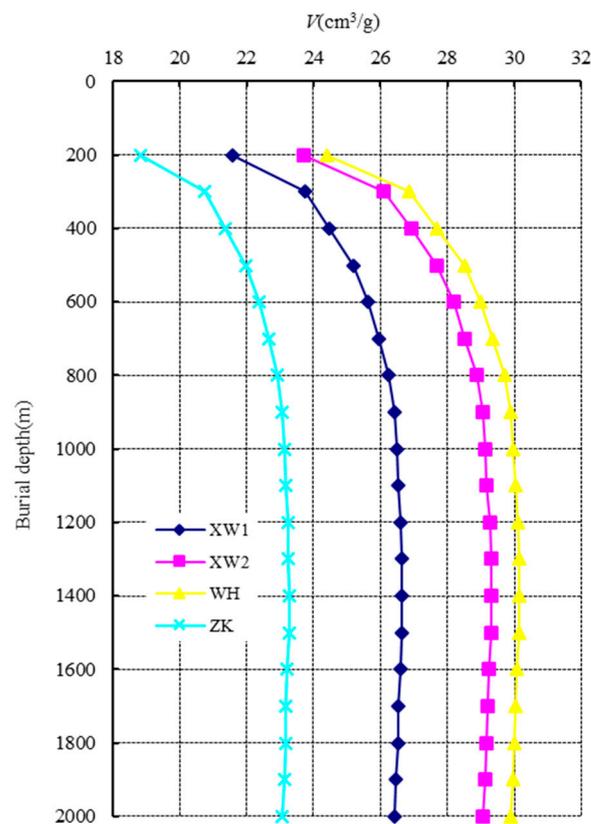


Figure 6. Predicted relationship between the methane adsorption capacity and the burial depth.

As shown in Figure 6, the amount of methane adsorbed by coal first increased rapidly with the increase in burial depth, tended to plateau to reach its maximum value at a depth

of approximately 1400 m, and then slowly decreased with a further increase in burial depth. This indicates that the influence of pressure was more important in methane adsorption than that of temperature in shallow coal seams. Furthermore, with the increase in burial depth, the control effect of the temperature on the adsorption capacity was more significant. The predicted results are consistent with the methane adsorption characteristics of the deep coal reservoirs [14]. Due to the small geothermal gradient, the negative effect of the adsorption temperature on adsorption does not fluctuate significantly with the change in burial depth. This is the difference in the adsorption characteristics of deep coalbed methane in the southern Sichuan region and certain other regions.

5. Conclusions

- (1) Four coal samples were collected from the southern Sichuan region (China) for methane isothermal adsorption tests at 30 °C. The results showed that the methane adsorption capacity of coal first increased rapidly with adsorption pressure and then gradually stabilized. The methane isothermal adsorption curve followed the Type I isothermal adsorption curve.
- (2) Four methane adsorption characteristic curves and corresponding equations were established and fitted based on the adsorption potential theory. The fitting results had a high correlation coefficient.
- (3) The methane isothermal adsorption curves of the four coal samples at 45, 60, and 75 °C were predicted based on the adsorption potential theory. It was found that the methane adsorption curves of coal under different temperature conditions belonged to the Type I isothermal adsorption group. The methane adsorption capacity on coal gradually weakened with the increase in adsorption temperature.
- (4) The adsorbed gas content in the reservoir first increased rapidly with the increase in the coal seam burial depth, reached a maximum value in the region, and then tended to decrease, indicating that the methane adsorption capacity was more significantly controlled by the pressure in shallow coal seams, whereas it was controlled by the temperature in deep coal seams.

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