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Research Article

Study on the Effect of Salinity and Water Content on CBM Adsorption/Desorption Characteristics of Coal Reservoir in Baode Block

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The adsorption/desorption characteristics of a coal reservoir play an important role in coalbed methane (CBM) development. The proximate analysis, maceral analysis and methane isothermal adsorption/desorption experiment are carried out based on coal samples from no. 4+5 coal seam in Baode block. Combing with coal experimental data and the CBM well-produced water salinity data in the Baode block, the effect of salinity and water content on CBM adsorption/desorption characteristics of the coal reservoir and its influencing mechanism is discussed. The results show that the CBM adsorption and desorption capacity decreases with the increase of water salinity and the decrease value shows a decreasing trend. The increase of water salinity reduces the solubility of methane in coal seam water and then reduces the adsorption capacity of methane. With the increase of water content, the adsorption and desorption capacities of CBM decrease gradually. The CBM adsorption and desorption capacities decrease with the increase of water content in coal samples. The adsorption/desorption capacities of coal samples change rapidly in the low-water content stage and slowly in the high-water content stage. The competitive adsorption effect and water blocking effect between water and methane molecules are the main influencing mechanisms of water content on CBM adsorption/desorption. It can be seen that the salinity and water content will have a certain adverse impact on the adsorption and desorption capacity of CBM in the Baode block. The influence of the difference in water content and salinity in the coal seam cannot be ignored in reserve evaluation and productivity prediction of CBM. The continuous, stable, and effective drainage is one of the key factors to ensure the efficient development of CBM wells in the Baode block.

1. Introduction

China is rich in CBM resources, and the amount of CBM resources buried at a depth of 2000 m or shallower is 30.05×10^{12} m^3, accounting for 11.6% of the total global CBM resources, with huge development potential [1]. According to incomplete statistics, by the end of 2020, 19540 vertical CBM wells and 1677 horizontal CBM wells had been drilled in China, in which 12880 had been put into production [2]. The accumulative proven CBM geological reserves are 9302×10^8 m^3, and the national production of CBM in 2020 was 58.2×10^8 m^3 [2]. Committing to achieving peak carbon dioxide emissions before 2030 and achieving carbon neutrality before 2060, promoting the efficient development and utilization of CBM is of great significance to "carbon neutrality;" however, the unconventional characteristics of CBM mainly in the adsorbed state have always been one of the difficult issues in the CBM development field.

Adsorption/desorption characteristics are the basis for revealing the occurrence mechanism and efficient development of CBM. The adsorption/desorption property has a significant impact on the gas content in a coal reservoir and the productivity of a CBM well and is an important parameter for evaluating the development potential of a coal reservoir [3–5]. The adsorption/desorption characteristics of a coal reservoir are usually characterized by methane isothermal adsorption/desorption experiment and the Langmuir equation, but the curves and parameters measured by the national standard method are unable to fully reflect the influence of high-salinity water on the adsorption/desorption characteristics in the in-situ state of a coal reservoir.

Previous studies revealed that salinity inhibits the adsorption capacity of coal seams [6–9]. Liu et al. carried out simulation experiments on the adsorption capacity of lignite to CBM under different salinity conditions and found that the increase of salinity could lead to the decrease of adsorption capacity in a coal reservoir [6]. Wang et al. studied the adsorption capacity of a coal core under different salinity conditions and believed that high salinity will reduce the adsorption capacity [7]. Yi et al. studied the methane desorption rate of granular anthracite coal samples under the condition of water injection with different salinity and found that the salinity reduced the methane desorption rate in coal [8]. Wei et al. found that there was a limit value for the influence of water salinity on the coal adsorption capacity and the limit was about 10000 mg/L for long-flame coal from the southern margin of the Junggar Basin [9].

Compared with the salinity, the influence of water on the adsorption/desorption characteristics of CBM was more widely studied. Joubert et al. first studied American bituminous coal samples and found that the adsorption of methane by moisture is closely related to the properties of coal. Under the condition of saturated moisture, water will not further affect the adsorption of methane in coal [10]. Krooss et al. carried out the adsorption experiments of methane and CO_2 on multiple coal samples under dry and wet conditions and found that the adsorption capacity of methane will decrease by 25% when the water content increases by 1% [11]. Other researchers in China successively carried out isothermal adsorption experiments on coal samples under the condition of equilibrium water. They believe that the molecular force between water and coal is stronger than methane, which reduces the adsorption performance of coal [12–14]. In addition, in the process of CBM development, when the water-based fracturing fluid invades the nanopore structure of coal, it will inevitably affect the adsorption/desorption characteristics of the coal reservoir and then affect the production of CBM wells.

Although predecessors have recognized that salinity and water will affect the adsorption/desorption capacity of coal reservoirs, no relevant research has been carried out on the Baode block, which is a hot spot area of medium-low-rank CBM development in China. Due to the low degree of thermal evolution of coal in the Baode block, the adsorption capacity of a coal reservoir is weak and the gas content is low. Therefore, compared with medium- and high0-rank coal blocks, the influence of salinity and water content on the adsorption/desorption capacity of coal reservoirs is particularly significant in this block. It has a great impact on the accuracy of CBM reserve calculation and coal reservoir adsorption/desorption capacity evaluation. In view of the abovementioned problems, relying on the major national science and technology project of CBM, this study studies the influence of salinity and water content on the adsorption/desorption characteristics of coal reservoirs in the Baode block, in order to provide basic support for the deployment and optimization of efficient exploration and development scheme of CBM in this block.

2. Sampling Background and Experimental Methods

2.1. Geological Setting. In the administrative division, the Baode block is located in the northwest of Shanxi province, covering an area of 476.46 km^2. Structurally, it is located in the northern part on the eastern edge of the Ordos Basin (Figure 1). It is generally characterized by a monoclinal structure inclined to the west, with a stratigraphic dip angle of $5^\circ \sim 10^\circ$, and the fault structure is not very developed. Groundwater in the block mainly comes from atmospheric precipitation and lateral recharge of Ordovician limestone. It is generally runoff from east to west, and the hydrological environment is in the runoff weak runoff area.

The main coal-bearing strata in the Baode block are the Taiyuan Formation and Shanxi Formation. The no. 4+5 coal seam in the Shanxi Formation and the no. 8+9 coal seam in the Taiyuan Formation are the main coal seams for CBM development [15]. The single-layer thickness of the no. 4+5 coal seam is between 5 and 14.6 m, with an average of 7.6 m. The single-layer thickness of no. 8+9 is between 5 and 14.2 m, with an average of 10.2 m. The vitrinite reflectance of the coal in the Baode block ranges from 0.71% to 1.22%, belonging to highly volatile bituminous coal [16].

2.2. Sample Collection. The coal samples used in this study were collected from no. 4+5 coal samples of Wangjialing coal mine in the Baode block (Figure 1). The sampling was carried out in accordance with the methods for coal seam sampling (following Chinese National Standards GB/T482-2008). The macrolithotypes and heterogeneity of coal seams were fully considered in the sample collection, and the samples were highly representative. The samples were collected from the fresh coal face of the mine and then immediately put into the sampling bag and fasten to prevent pollution and oxidation.

2.3. Experimental Methods. The maximum vitrinite reflectance ($R_{o,max}$, %) measurements and maceral analysis were performed on the same polished section of coal samples, following Chinese National Standards GB/T6948-1998 and GB/T8899-1998. Proximate analysis was performed following Chinese National Standard GB/T212-2001 for samples to obtain the ash yield, moisture, and volatile contents of the coals.

The isothermal adsorption experiment is the main technical method to characterize the ability of a coal reservoir to adsorb CBM. In order to study the influence of salinity and water content on adsorption/desorption characteristics of coal reservoirs, methane isothermal adsorption desorption tests of coal samples from the Baode block under different

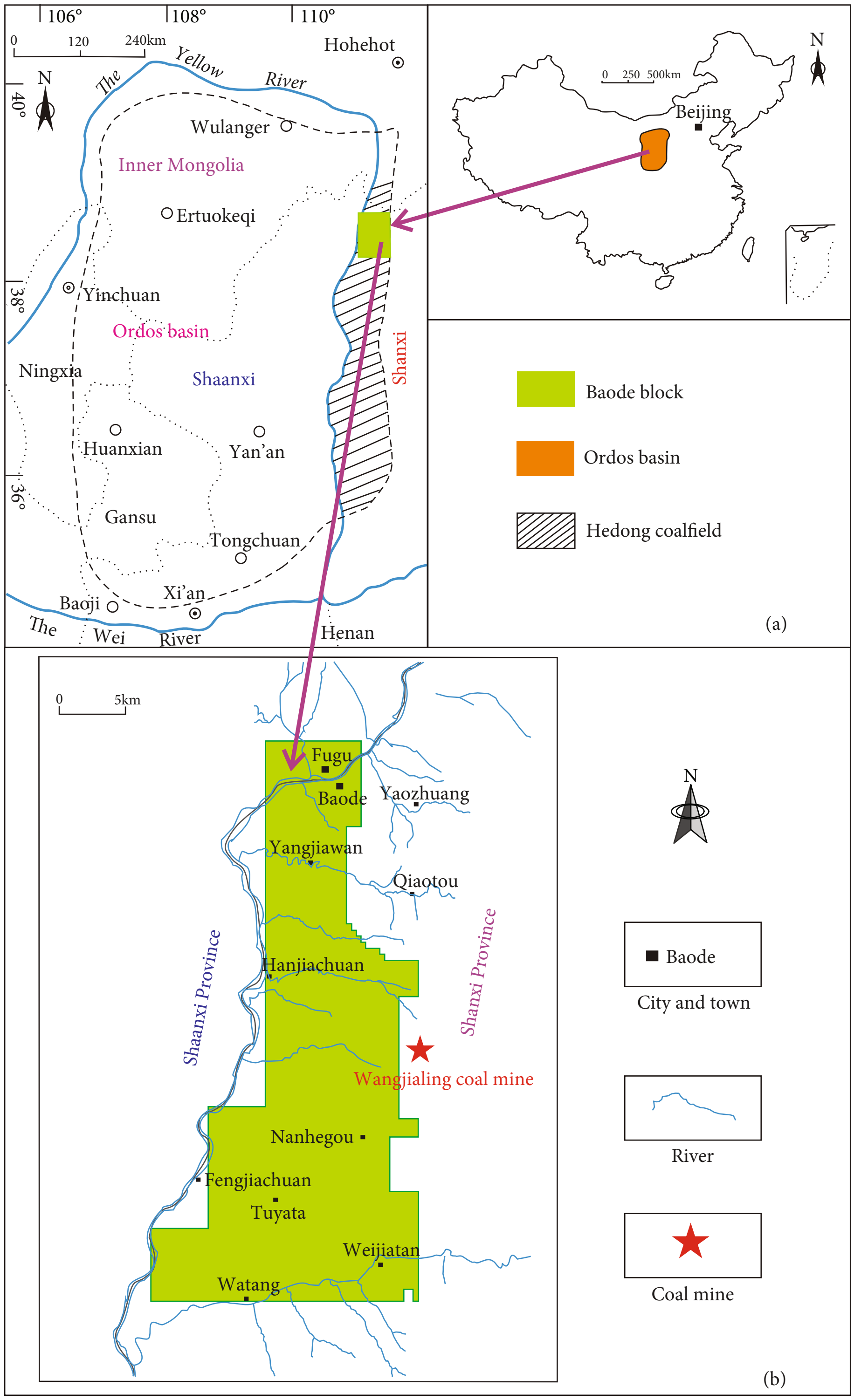

FIGURE 1: (a) Location of the Ordos Basin and Baode block. (b) Location of Wangjialing coal mine and the scope of the Baode block.

salinity and water contents were carried out. The experimental instrument adopts the automatic isothermal adsorption instrument, which is implemented in accordance with the high-pressure isothermal adsorption test method of coal (GB/T 19560-2008). It should be noted that all adsorption capacity data and Langmuir isothermal adsorption parameter data in this study are based on air drying basis. The experimental tests were carried out in the Shanxi Key Laboratory of Coal and Coal-Measure Gas Geology.

3. Results and Discussions

3.1. Coal Petrology and Quality Characteristics. The results of vitrinite reflectance, proximate analysis, and maceral

Table 1: Coal vitrinite reflectance, proximate analysis, and maceral composition in the Baode block.

Sample no.	$R_{o,max}$ (%)	Proximate analysis (%)			Maceral composition (%)			
		M_{ad}	A_d	V_{daf}	V	I	E	MM
BD-WJL-04	0.81	2.16	14.44	35.62	48.7	32.1	5.8	13.6

$R_{o,max}$: maximum vitrinite reflectance; V: vitrinite; I: inertinite; E: liptinite; MM: mineral matter; M_{ad}: moisture (air dry basis (ad)); A_d: ash yield (air dry basis (ad)); V_{daf}: volatiles (dry and ash free (daf)).

composition of the coal samples are shown in Table 1. The maximum vitrinite reflectance of the experimental coal sample is 0.81%, belonging to highly volatile bituminous coal. The organic maceral composition of the experimental coal sample is dominated by vitrinite (48.7%), followed by inertinite (32.1%) and liptinite(5.8%). Besides, the proximate analysis result shows that moisture, ash yield, and volatiles are 2.16%, 14.44%, and 35.62%, respectively.

3.2. Effects of Salinity on Adsorption/Desorption CBM Characteristics. This study carried out the experimental analysis of methane isothermal adsorption and desorption of coal samples under different salinity conditions for no. 4+5 coal samples of Wangjialing coal mine. Since the coal seam water salinity measured by the chemical analysis of the produced water of no. 4+5 CBM well in the Shanxi Formation of the Baode block ranges from 902 to 2026 mg/L (Table 2) [17], the salinity gradients of the simulated formation water used in the isothermal adsorption/desorption experiment were set as 1000, 1400, 1800, and 2200 mg/L in this study.

3.2.1. Effects of Salinity on CBM Adsorption/Desorption Capacity. By comparing the adsorption and desorption curves of coal samples from the Baode block soaked in water with different salinity gradients (1000, 1400, 1800, and 2200 mg/L), it can be found that the adsorption and desorption capacities of CBM generally decrease with the increase of coalbed water salinity and the decline value of adsorption and desorption capacities generally decrease with the increase of coalbed water salinity (Figure 2).

The isothermal adsorption constants of the coal samples with different salinity gradients (1000, 1400, 1800, and 2200 mg/L) were calculated by the Langmuir equation (Table 3). The Langmuir volumes of the coal samples were 18.75, 15.68, 13.86, and 12.43 m^3/t, and the Langmuir pressure values are 3.86, 3.18, 3.08, and 2.79 MPa, respectively. The adsorption capacity with a salinity of 1000 mg/L is the largest. The higher the salinity, the smaller the Langmuir volume characterizing the adsorption capacity, indicating that the salinity of coalbed water will have an adverse impact on the adsorption of CBM.

3.2.2. Effects of Different Salinity on CBM Desorption Lag Characteristics. The isothermal adsorption/desorption curves of no. 4+5 coal samples from the Baode block under different salinity conditions are shown in Figure 2. It can be found that for each salinity gradient, there is always a desorption lag in the desorption curve, reflecting that the adsorption capacity of CBM is greater than the desorption capacity under the same differential pressure. From the comparison of curve characteristics under different salinity, it is evident that the desorption lag phenomenon has no obvious relationship with the salinity gradients (Figure 2). Because the equivalent adsorption heat of CBM desorption is greater than that of adsorption, the desorption process needs to absorb heat from the outside system. The energy difference between the adsorption process and the desorption process may be the key factor of the CBM desorption lag [18]. Ma et al. analyzed from the perspective of the pore structure and believed that the main reason for the desorption lag was the stronger binding ability of micropores and small pores to methane molecules [19].

3.2.3. Effect Mechanism of Salinity on CBM Adsorption/Desorption Characteristics. Wei et al. studied the massive coal samples of the Jurassic Xishanyao Formation in the Manas mining area on the southern edge of the Junggar Basin; they found that with the increase of inorganic salt concentration in the solution, the surface tension of the solution increased and the capillary phenomenon became more obvious, which further reduced the adsorption site on the surface of the coal matrix, thus reducing the Langmuir volume and adsorption capacity of coal samples to methane. However, there is also a limit value. If the salinity is greater than 20000 mg/L, the salinity can no longer significantly reduce the Langmuir volume of coal samples [9].

In recent years, with the successful application of nanopore analysis and molecular simulation technology in the field of unconventional oil and gas, some scholars have verified and explained the characteristics of methane adsorption/desorption from the perspective of numerical simulation. Zhou et al. confirmed through the research of molecular simulation technology that salinity has a negative impact on the methane adsorption capacity of kerogen [20]. In a certain salinity range, the higher the salinity, the greater the impact on methane adsorption, and when the salinity was 6 mol/L and the temperature was 338.15 K, the methane adsorption capacity decreased by about 6.0% [20] (Figure 3). The existence of NaCl reduces the solubility of methane in brine and then reduces the adsorption capacity of methane. These research results can also explain the reason why the methane adsorption capacity and desorption capacity of the coal reservoir in the Baode block decrease with the increase of coalbed water salinity in this study from aspects of experiment and numerical simulation. Meanwhile, it also indicates that the influence of salinity should not be ignored in the evaluation of the CBM reservoir.

3.3. Effects of Water Content on CBM Adsorption/Desorption Characteristics. This study carried out the experimental analysis of methane isothermal adsorption and desorption

Table 2: The ion composition and water type of produced water in no. 4+5 coal in the Baode block. [17].

Well no.	Cationic concentration (mg/L)			Anion concentration (mg/L)			Salinity (mg/L)	Water type
	$Na^+ + K^+$	Mg^{2+}	Ca^{2+}	SO_4^{2-}	Cl^-	HCO_{3-}		
B-1	283	44	29	7	336	471	1175	$NaHCO_3$
B-2	398	69	59	21	558	474	1580	$NaHCO_3$
B-3	305	43	51	7	319	465	1190	$NaHCO_3$
B-8	432	50	75	1	591	460	1610	$NaHCO_3$
B-11	577	57	64	8	607	715	2026	$NaHCO_3$
B-12	252	24	36	4	236	781	1333	$NaHCO_3$
B-23	128	45	25	82	114	311	707	$NaHCO_3$
B-22	105	22	30	2	130	432	721	$NaHCO_3$
B-24	314	63	64	23	425	413	1301	$NaHCO_3$
B-25	287	41	22	42	282	393	1067	$NaHCO_3$
B-26	167	73	50	189	243	180	902	Na_2SO_4
B-27	459	16	24	10	460	567	1538	$NaHCO_3$
B-29	174	51	91	98	198	432	1045	Na_2SO_4
B-17	574.6	54.7	125	1	725	485	1969	$NaHCO_3$
B-19	604	53.4	25	3	733	522	1943	$NaHCO_3$

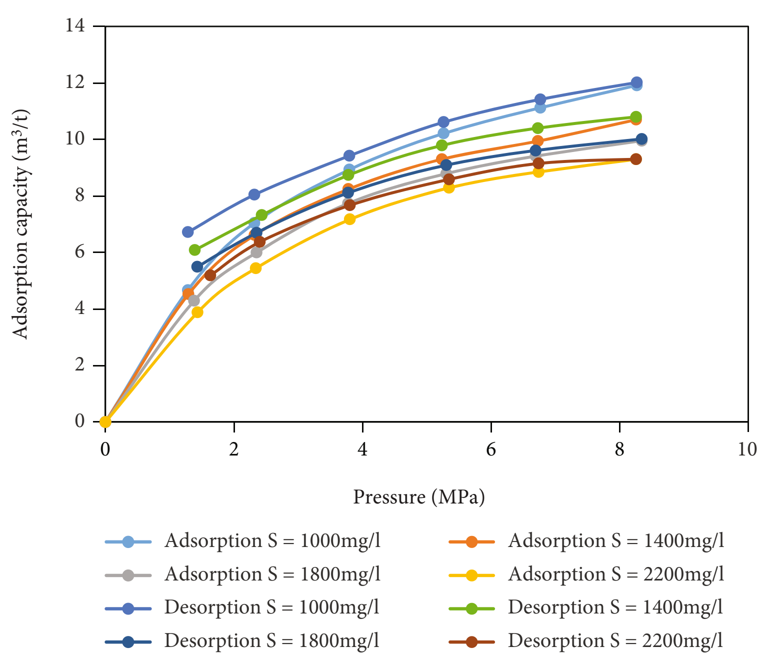

Figure 2: Methane isothermal adsorption/desorption curves of coal samples with different salinity.

Table 3: Langmuir isothermal adsorption parameters of Baode coal samples with different salinity.

Salinity gradients	V_L (m^3/t)	P_L (MPa)
1000 mg/L	18.75	3.86
1400 mg/L	15.68	3.18
1800 mg/L	13.86	3.08
2200 mg/L	12.43	2.79

of coal samples under different water content gradients for no. 4+5 coal samples of Wangjialing coal mine. The water content gradient values are set according to the equilibrium water content value of the no. 4+5 coal seam in the Baode block. The preliminary exploration well coal core experiment shows that the equilibrium water saturation obtained from the isothermal adsorption experiment of lump coal samples in Baode district is about 5%. Therefore, in this study, the water content gradients of experimental coal samples are set as 2%, 3%, 4%, and 5%.

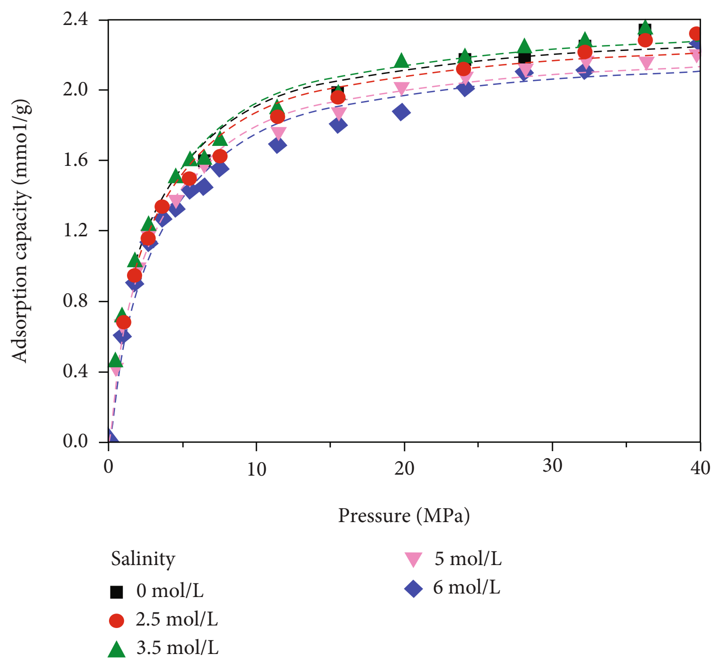

FIGURE 3: Methane adsorption isotherms of different salinities with the water content of 3 wt % at 338.15 K. Points are results of simulation, and lines are results of Langmuir fitting [20].

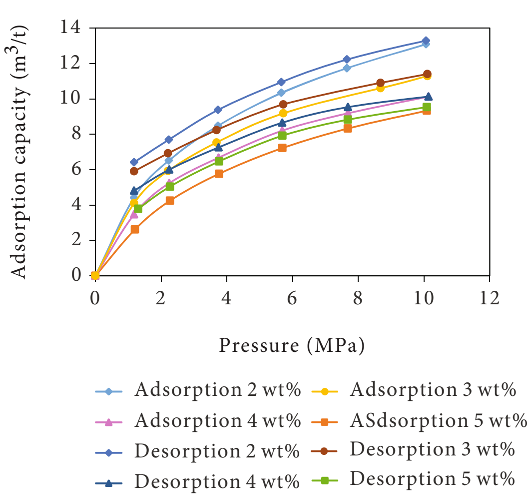

FIGURE 4: Methane isothermal adsorption/desorption curves of coal with different water contents.

TABLE 4: Langmuir isothermal adsorption parameters of Baode coal samples with different water content.

Coal samples with different water content	V_L (m^3/t)	P_L (MPa)
2%	16.78	3.52
3%	14.09	3.05
4%	13.05	3.35
5%	13.03	4.71

3.3.1. Effects of Water Content on CBM Adsorption/Desorption Capacity. By comparing the adsorption and desorption curves of coal samples with different water contents of coal 4+5 in the Baode block, it can be found that the CBM adsorption and desorption capacities generally decrease with the increase of water content in coal samples (Figure 4). The CBM adsorption and desorption in coal samples will decrease with the increase of water content. In the stage of low water content, the adsorption/desorption of methane changes rapidly, and in the stage of high water content, the change is slow. It can be seen that the high water content of coal seams will have an adverse impact on the rapid and efficient desorption of CBM in the Baode block.

The isothermal adsorption constants of the samples were calculated by the Langmuir equation (Table 4). The Langmuir volumes of the samples were 16.78, 14.09, 13.05, and 13.03 m^3/t, and the Langmuir pressure values are 3.52, 3.05, 3.35, and 4.71 MPa, respectively. The adsorption capacity with a water content of 2% is the largest. The higher the water content, the smaller the Langmuir volume characterizing the adsorption capacity, indicating that the water in the coal seam will have an adverse impact on the desorption/adsorption of the CBM.

3.3.2. Effects of Water Content on CBM Desorption Lag Characteristics. The isothermal adsorption/desorption curve of no.4+5 coal samples of the Shanxi Formation in the Baode block under different water content conditions is shown in Figure 5. It can be seen that there is always a desorption lag in coal samples under different water content conditions but there is no obvious correlation with the water content (Figure 4). Previous studies have shown that the lag mechanism of coal samples is mainly reflected in two aspects, i.e.,

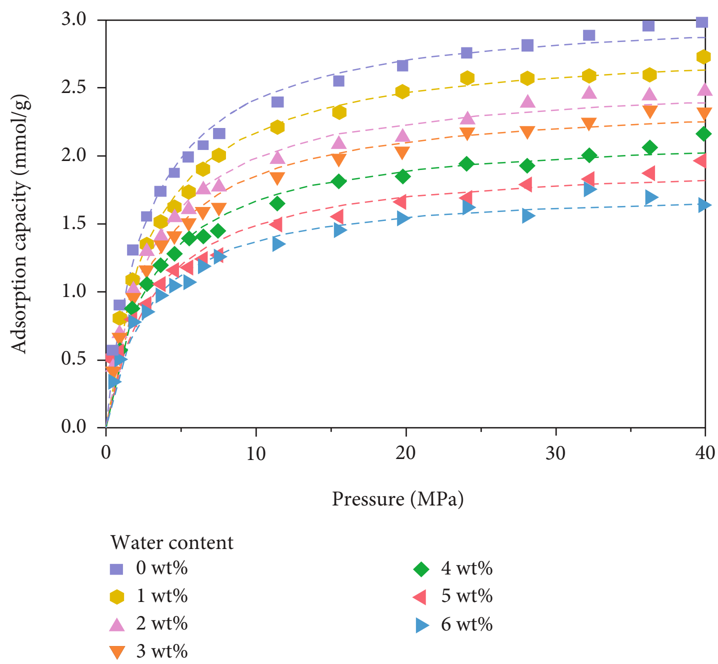

Figure 5: CH_4 adsorption isotherms under different water contents at 338.15 K. Points are results of simulation, and lines are results of Langmuir fitting [20].

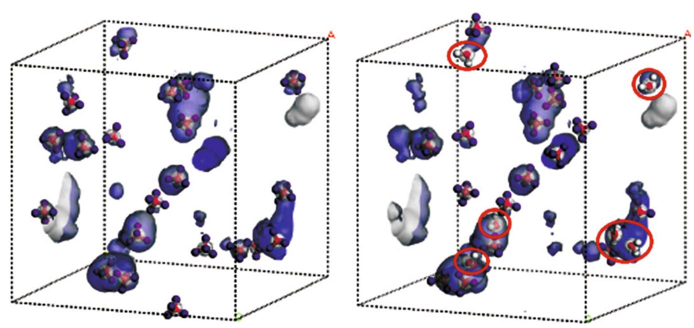

Figure 6: Simulation of adsorption of CH_4 on coal macromolecules with different water content [25] (red elliptic circles represent water competitive adsorption sites).

desorption obstruction and gas migration obstruction. The moisture is one of the main factors affecting the desorption lag of CBM. The lag degree of the water-bearing coal sample is significantly higher than that of the dry coal sample, but it has no obvious relationship with water content [21, 22], which is confirmed by the results of this study.

3.3.3. Effect Mechanism of Water Content on CBM Adsorption/Desorption Characteristics. Previous studies have shown that water has a significant impact on the adsorption/desorption characteristics of the CBM [12–14, 21, 22]. The influence mechanism of water content on methane adsorption/desorption characteristics of coal samples is mainly reflected in two aspects, i.e., the competitive adsorption effect between water and methane molecules and the water lock effect.

Because the interaction force between coal and water molecules is much greater than that of coal and methane molecules, in the case of three-phase medium coexistence, water and methane molecules produce competitive adsorption on the coal surface and water molecules will be replaced to partially adsorb methane, resulting in the reduction of methane adsorption capacity of the coal reservoir. Zhou et al. confirmed through molecular simulation technology that the increase of water content will reduce the adsorption capacity of the kerogen to methane (Figure 5). The density distribution changes of methane and water molecules show that water molecules occupy some adsorption sites in the kerogen matrix to prevent methane from entering the nanopores, which leads to the decline of methane adsorption capacity of the kerogen [20].

The water content of coal is closely related to the coal wettability, which can indirectly affect the adsorption/desorption characteristics of CBM [23, 24]. Water molecules are easy to combine with the broken chemical bonds on the surface of the coal matrix and the hydrophilic functional

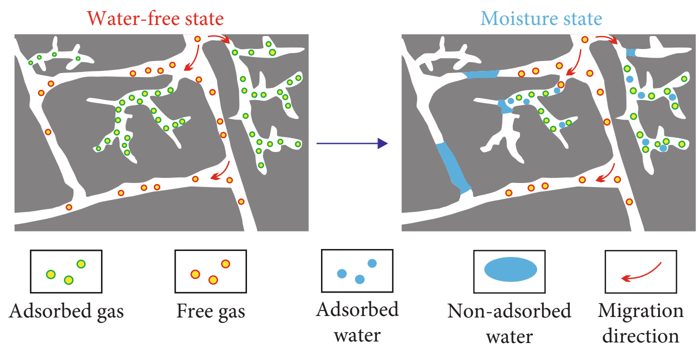

FIGURE 7: Influence of water locking effect on adsorption/desorption characteristics of methane in coal (modified from literature [26]).

groups inside the coal matrix, resulting in the enhancement of hydrophilicity of the coal. The surface free energy of coal is reduced to a certain extent, and the heat released by reaching the methane-coal adsorption system equilibrium state is decreasing. The molecular force between water and coal is stronger than that of methane, which can occupy effective adsorption sites on the coal surface and weaken the methane adsorption capacity of coal (Figure 6) [25].

In addition, the continuous accumulation of adsorption of water molecules on the surface of micropores and fissures in coal will lead to water molecules occupying some pore and throat channels in coal, leading to a water lock effect [26]. The higher the water content of the coal is, the stronger the water locking effect is. The existence of the water lock effect leads to the blockage of some pore channels and the obstruction of methane movement channels, which leads to the decrease of methane adsorption and desorption capacity in coal (Figure 7). In this study, the experimental results of methane adsorption and desorption capacities of coal reservoir in the Baode block decrease with the increase of coal water content, which is consistent with the understanding of previous theoretical studies and also indicates that the influence of coal water content cannot be ignored in the reserve evaluation and development effect prediction of CBM in a high water-bearing coal seam.

4. Conclusion

(1) Desorption lag phenomenon exists, under any condition of salinity and water content, in coal of the Baode block, but there are differences in desorption lag degree with different salinity and water content. The moisture content is the main factor affecting the desorption lag. The CBM desorption lag mechanism is mainly reflected in blocked desorption and blocked gas migration. The energy difference between the adsorption and desorption processes is the key internal factor of the desorption lag. The influence of water content and the strong binding effect of micropores on methane are also important factors of the CBM desorption lag

(2) With the increase of salinity, the adsorption and desorption capacities of coal in the Baode block show a decreasing trend in general and the decrease value of adsorption and desorption capacities shows a decreasing trend. The increase of coal seam water salinity reduces the solubility of methane in brine and then reduces the adsorption capacity of methane. The influence of coalbed water salinity cannot be ignored in the reserve evaluation of CBM resources

(3) The adsorption and desorption capacities of CBM decrease gradually with the increase of water content. The adsorption and desorption capacities decrease rapidly in the stage of low water content, while slowly in the stage of high water content. The competitive adsorption effect and water locking effect between water and methane molecules are the main influencing mechanisms of water content on methane adsorption/desorption. The influence of water content in the coal seam cannot be ignored in CBM reserve evaluation and productivity prediction in a high water-bearing coal seam

(4) The salinity and water content in the coal reservoir will have a certain adverse effect on the adsorption and desorption capacities of CBM in the Baode block. Continuous, stable, and effective water production is one of the key factors to ensure the efficient development of CBM in this block

Data Availability

All data are from our team's coal sample experimental test data and references, which have been described in detail in this paper.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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