Effect of Electric Potential Gradient on Methane Adsorption and Desorption Behaviors in Lean Coal by Electrochemical Modification: Implications for Coalbed Methane Development of Dongqu Mining, China

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ABSTRACT: The application of electrochemical modification for accelerating methane extraction in lean coal seams is limited due to the lack of experimental and theoretical research studies. Therefore, electrochemical modification with different electric potential gradient values was selected to modify lean coals in this study; meanwhile, the amount of methane adsorption and the methane desorption ratio were tested and analyzed. The results showed that the maximum amount of methane adsorption in coal samples decreased after electrochemical modification and the decrease in methane adsorption increased with an increase in electric potential gradient. The methane desorption ratio increased from 83.20% up to 87.84 and 86.90% at the anode and cathode zone, respectively, after electrochemical modification using a 4 V/cm electric potential gradient. A higher electric potential gradient performs better in the electrochemical modification. The mechanism of electrochemical modification using different electric potential gradients was revealed based on the measurements of Fourier transform infrared spectroscopy and liquid nitrogen adsorption. It is due to an increase in acid groups in coal molecular structure and the change of the specific surface area of coal after modification. The results obtained from this work contribute to the methane extraction via the electrochemical method in lean coal seams.

1. INTRODUCTION

Extracting coalbed methane (CBM) from the underground has attracted global attention because of its significance to clean energy supplies. Previous studies showed that more than 80% of CBM exists in the coal seam as an adsorbed state. The adsorption and desorption behavior of CH₄ on the coal surface is closely related to the composition, pore structure, and surface characteristics of coal samples. Lean coal in Dongqu Coal Mine of the Xishan coalfield in China belongs to low-volatility high-rank bituminous coals with high inner surface area and absorbs a large amount of methane. Therefore, it is urgent to study the mechanism of adsorption and desorption of the lean coal modified by an electrochemical method, which could improve the efficiency of CBM extraction.

Physical and chemical modifications have been studied to analyze their impact on methane adsorption and desorption in coal and coal-based activated carbons. Li et al. found that methane adsorption was enhanced in raw coal and tectonically deformed coal under an electrostatic field. Lean coal in Dongqu Coal Mine of the Xishan coalfield in China belongs to low-volatility high-rank bituminous coals with high inner surface area and adsors a large amount of methane. Therefore, it is urgent to study the mechanism of adsorption and desorption of the lean coal modified by an electrochemical method, which could improve the efficiency of CBM extraction. Li et al. found that methane adsorption was enhanced in raw coal and tectonically deformed coal under an electrostatic field. Acid modification can increase the content of oxygen and functional groups on the coal surface, which leads to a decrease in methane adsorption. Feng et al. utilized H₂SO₄ and (NH₄)₂S₂O₈ to modify bituminous coal, and the experimental results showed that the O-containing groups over the coal surface increased while the methane adsorption decreased. The acid modification (H₂SO₄ and HNO₃) of coal-based activated carbons leading to lower methane adsorption has been reported by Zhang et al. In contrast to acid modification, an alkaline solution-modified coal showed a stronger adsorption capacity for methane; the maximum adsorption capacity of bituminous coal increased from 23.99 to 27.52 mL/g and increased from 33.62 to 39.94 mL/g for anthracite after the alkaline modification. In the electrochemical modification method, the acidic and alkaline treatments on coal samples were conducted in the anode and cathode environment, respectively, in which the electrokinetic characteristics of coal were changed.
When the coal particles are immersed in solution, the carboxyl and aldehyde functional groups on the surface of the coal are dissociated, and the ions in the solution are adsorbed onto the coal surface, which will lead to the formation of an electric double layer on the surface of the coal.\(^1\) Coal particles are electronegative due to dissociation, while the metal ions from minerals are positively charged; so, they move to the positive and negative electrode zones, respectively, under an external direct current (DC) electric field.\(^5\),\(^11\)

Besides, the capacity of methane adsorption in coal could be reduced, while the methane desorption ratio could be increased by the electrochemical modification. Thus, the extraction efficiency of CBM can be improved. Hydrogen ions and electrolysis heat generated in the electrolysis process have a significant impact on the adsorption capacity of methane in coal.\(^6\),\(^12\) Zhang et al.\(^6\) studied the adsorption characteristics of coal-based activated carbon modified by different acids and put forward that the inhibition of methane adsorption by an increase in acid functional groups after modification is stronger than the promotion of pore structure changes. There is a negative correlation between temperature and methane adsorption capacity.\(^12\) Kang\(^13\) found that electrochemical modification had a significant effect on the methane adsorption capacity of anthracite, and the adsorption capacity after modification was only 48.63% of that before modification. During the electrochemical modification, oxidation, reduction, and electroosmosis promote the desulfurization of coal and the removal of mineral matters, which contribute to the formation of new pores and fractures in coal and the extension and connection of existing pores and fractures.\(^9\),\(^14–16\) Guo et al.\(^9\) found that the minerals in the cracks about 90 \(\mu\)m of coal disappeared after electrochemical modification, which resulted in a 68% reduction in desorption equilibrium time of modified samples. The sulfides in fossil fuels could be removed by electrochemical redox reaction through an electrochemical desulfurization method (ECDS). At the anode, the oxidation product, whose polarity was enhanced, can be extracted with polar solvents or polar adsorbents, while in the cathode region, the reduction product can be extracted with hydrogen sulfide. In addition to the electrolyte and electrode materials, the potential gradient also plays an important role in the effect of electrochemical modification. It is generally reported that the electrochemical modification including the rate of electro-oxidation,\(^17\) dewatering of sludge,\(^18\) and gas desorption\(^1\) has a better effect with an increase in potential gradient.

In summary, scholars have done lots of works to study the effect of modification on coal for desulfurization, dewatering, and methane adsorption/desorption by an electrochemical method. However, the research studies about the effect of electric potential gradient on methane adsorption/desorption in lean coals modified by an electrochemical method are limited. The logging data show that lean coal reservoirs contain huge amounts of methane in Donggu Mining of China, while the efficiency of CBM extraction is extremely low. Thus, accelerating CBM extraction via the electrochemical method still needs further research. The effect of electric potential gradient on CH\(_4\) adsorption/desorption in modified coal needs more explanation. In this paper, the lean coal was modified by an electrochemical method at four different potential gradients, namely, 0, 1, 2, and 4 V/cm, respectively. The adsorption/desorption behavior, the reduction of surface free energy, surface oxygenic groups, and pore size distribution of original and modified coal samples were measured and analyzed to reveal the effect and mechanism of electrochemical modification with different potential gradients.

2. RESULTS AND DISCUSSION

2.1. CH\(_4\) Adsorption Results after Modification. The test results of methane adsorption on the coal samples before and after electrochemical modification are shown in Figure 1, and the methane adsorption data was analyzed by the Langmuir model.\(^19\)

\[
V_{ab} = \frac{V_lP}{P_l + P}
\]

where \(V_{ab}\) is the amount of adsorbed gas (mL/g); \(V_l\) is the maximum amount of adsorbed gas (mL/g); \(P_l\) is the gas equilibrium pressure of half of the saturated adsorption amount \(V_l\); \(V_l\) and \(P_l\) are called Langmuir constants, and the fitting results are listed in Table 1.

The data in Figure 1 show that methane adsorption of lean coal decreased after electrochemical modification using a Na\(_2\)SO\(_4\) electrolyte at different electric potential gradients. The adsorption capacity at both the anode and cathode decreased in the order of T1 > T2 > T3 > T4, which means that the

![Figure 1. Methane adsorption of lean coal samples with pressure at 293.15 K. (a) Anode zone. (b) Cathode zone.](https://dx.doi.org/10.1021/acsomega.0c03496)
effect of electrochemical modification performs better at a higher potential gradient. Table 1 shows that the maximum adsorption amount $V_L$ in unmodified lean coal was 30.030 mL/g, which decreased respectively to 29.239 and 29.851 mL/g at the anode and cathode after modification using a 1 V/cm potential gradient. Then, the $V_L$ values further decreased to 28.329 and 28.653 mL/g, respectively, after modification using a 2 V/cm potential gradient. The maximum adsorption amount of methane decreased to 26.667 and 27.248 mL/g after electrochemical modification using a 4 V/cm potential gradient, which is the best modification condition to decrease the amount of methane adsorbed on lean coal. The Langmuir constant $P_L$ varied in the same trend with the potential gradient (listed in Table 1). $P_L$ can be used to evaluate the affinity of coal to methane and the ability of methane desorption under reservoir pressure.\(^{20,21}\) The experimental fitting results showed that $P_L$ increases with an increase in potential gradient, illustrating that methane adsorption occurs more difficulty and desorption is more available at a higher potential gradient.

When methane was adsorbed on the lean coal surface, the surface excess $\Gamma$ could be expressed as\(^{22}\)

$$\Gamma = \frac{V}{V_0S}$$

(2)

where $V$ is the amount of adsorbed methane (L), $V_0$ is the methane volume (L/mol), and $S$ is the surface area (m$^2$/g).

The Gibbs equation could be expressed as

$$-d\gamma = RT\Gamma d(ln P)$$

(3)

where $\gamma$ represents surface tension (J/m$^2$), $R$ represents the universal gas constant, $T$ is the absolute temperature (K), and $P$ is the absolute methane pressure (Pa). Combining eqs 2 and 3, the equation can be written as

$$\Delta\gamma = \gamma_0 - \gamma_1 = \frac{RT}{V_0S} \int_0^\infty \frac{V}{P} dP$$

(4)

where $\gamma_0$ is the surface tension without methane adsorption (J/m$^2$), and $\gamma_1$ represents the surface tension when methane is adsorbed (J/m$^2$).

Set $V_L = a$, $1/P_L = b$, and introducing eq 1 into eq 4, the following results are obtained

$$\Delta\gamma = \frac{RT}{V_0S} \int_0^\infty \frac{ab}{1 + bP} dP = \frac{aRT}{V_0S} \ln(1 + bP)$$

(5)

Equation 5 shows that the $\Delta\gamma$ of lean coal samples were relevant to the methane adsorption, the surface area of lean coal samples, the temperature, and the pressure when methane is adsorbed. The calculation results of the $\Delta\gamma$ of lean coal samples are shown in Figure 2, which shows that the $\Delta\gamma$ increased after methane is adsorbed, while the $\Delta\gamma$ decreased after modification, and the decrease in $\Delta\gamma$ increased with an increase in potential gradient up to 4 V/cm. Besides, the coal samples from the anode zone (Figure 2a) decreased larger than coal samples from the cathode zone (Figure 2b) at the same electric potential gradient, and the same results were also found by ref 23. The pressure and temperature of methane adsorption tests were the same; so, the reduction of coal surface energy can only be caused by electrochemical modification.

2.2. CH$_4$ Desorption Results after Modification.

Desorption results were characterized by the desorption ratio (%); the equation is as follows

$$\text{Desorption ratio} = \frac{\text{Amount of methane desorbed}}{\text{Amount of methane adsorbed}} \times 100\%$$

Figure 2. Reduction of surface free energy after electrochemical modification. (a) Anode zone. (b) Cathode zone.

Figure 3. Methane desorption ratio of lean coal samples after modification with different electric potential gradients. (a) Anode zone. (b) Cathode zone.
The desorption ratio $P$ in lean coal at the anode and cathode zone are shown in Figure 3. The desorption ratio data agree with the calculation results of methane desorption ratio increased to 87.840 and 86.900%, respectively, after electrochemical modification using a 1 V/cm electric potential gradient and further increased to 85.541 and 84.9796% after electrochemical modification using a 2 V/cm electric potential gradient. When the electric potential gradient increased to 4 V/cm, the methane desorption ratio increased to 87.840 and 86.900%. The desorption ratio data agree with the calculation results of $P_i$ in methane adsorption.

The diffusion coefficient can be used to characterize the CH$_4$ desorption capacity of coal.$^{24}$ The desorption expression is

$$\frac{Q_t}{Q_\infty} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(-\frac{n^2 \pi^2 D}{r_0^2} t\right)$$

(7)

where $Q_t$ represents the accumulative total methane diffusion during the time $t$ (cm$^3$/g); $Q_t/Q_\infty$ represents the accumulative total methane diffusion rate during the time $t$; $D$ represents the diffusion coefficient (cm$^2$/s); $r_0$ represents the radius of samples (cm); $t$ represents the desorption time (s). In order to simplify the calculation, Shi et al.$^{25}$ let $n = 1$, and then, eq 7 can be expressed as

$$\ln\left(1 - \frac{Q_t}{Q_\infty}\right) = -\lambda t + A$$

(8)

where $\lambda = \pi^2 D/r_0^2$, $A = \ln(6/\pi^2)$. $-\lambda$ and $A$ are the slope and the intercept of the $\ln(1 - Q_t/Q_\infty)$-$t$ curve. Based on the desorption data from Figure 3 and eq 8, the calculation results of the diffusion coefficient are shown in Figure 4. The calculation results show that the $D$ of unmodified lean coal is $7.099 \times 10^{-10}$ cm$^2$/s, increased to $8.113 \times 10^{-10}$ and $7.827 \times 10^{-10}$ cm$^2$/s after electrochemical modification using a 1 V/cm electric potential gradient and further increased to $9.289 \times 10^{-10}$ and $8.905 \times 10^{-10}$ cm$^2$/s after modification using a 2 V/cm electric potential gradient. The diffusion coefficient $D$ increased to $10.1424 \times 10^{-10}$ and $9.389 \times 10^{-10}$ cm$^2$/s after electrochemical modification using a 4 V/cm electric potential gradient. The increase in the $D$ indicated that the methane desorption after electrochemical modification was improved via a Na$_2$SO$_4$ electrolyte at 1, 2, and 4 V/cm electric potential gradients. The $D$ of the anode zone was higher than that of the cathode zone, indicating that the anode zone is more ready for desorption of methane, which was consistent with the results of Figure 3a,b.

2.3. Change of Functional Groups after Electrochemical Modification. The FTIR results of lean coal samples at the anode and cathode zone are shown in Figure 5. The peak near 3800 cm$^{-1}$ was due to the stretching of NH ($-\text{NH}$ and $-\text{NH}_2$).$^{26}$ At 507 and 1082 cm$^{-1}$, the peak of the smectite mineral was greatly reduced. Feng et al.$^{4}$ found that mineral matters in coal samples preferentially adsorbed methane due to their smaller mesostructures than the coal matrix. The reduction of the clay minerals reduced the adsorption of methane, which was consistent with the adsorption test results in Figure 1. The peaks near 876 and 1430 cm$^{-1}$ decreased, which indicates that the content of CaCO$_3$ in lean coals decreased. Guo et al.$^8$ pointed out that this result occurs due to the reaction of H$^+$ generated by anode reactions with CaCO$_3$. At the wavenumber around 1600 cm$^{-1}$, the peak represents C=O stretching, which can be provided by O-containing groups near the 1500–1800 cm$^{-1}$ wavenumber.$^{27}$ After electrochemical modification, O-containing groups around 1600 cm$^{-1}$ decreased as shown in T2, T3, and T4 of Figure 5, which verified the adsorption results shown in Figure 1.

2.4. Change of Mesopores after Electrochemical Modification. Pore structure is one of the main factors affecting the adsorption and desorption of methane in coal, especially the micropore with a pore size less than 10 nm. Figure 6 shows the distribution of pore size of coal samples at the anode and cathode zone. Tables 2 and 3 show the test results of pore characteristics from the anode zone and cathode zone, respectively. In the anode zone, the surface area of coal samples decreased first and then increased, and the pore volume of coal samples increased gradually with an increase in potential gradient of electrochemical modification. Compared with 3.3637 m$^2$/g of T1, the surface area of T2, T3, and T4 decreased or increased to 2.8917, 4.0727, and 6.6789 m$^2$/g; both the decrease and increase were mainly due to the contribution of pores with a pore size less than 10 nm. The pore volume of T2, T3, and T4 increased from 0.004249 cm$^3$/g of T1 to 0.004677, 0.005276, and 0.008932 cm$^3$/g. The pore volume of T2 and T3 with a pore size less than 3 nm was smaller than that of T1, and the other pore size ranges were larger than that of T1. The pore volume of T4 was larger than that of T1 in any pore size range. It indicated that pores with a pore size less than 10 nm are the main contributors to the surface area and pore volume. Methane adsorption decreased while the surface area increased after electrochemical modification, which could be explained by the increase in functional groups. Zhang et al.$^{23}$ also found this phenomenon in their experiments and found that the change of functional groups on the coal surface dominated the modification. When Hao$^{25}$ et al. modified bituminous coal, they also found that when the number of micropores in the coal sample changed little, the adsorption capacity of methane in the coal sample with high oxygen content and poor hydrophobicity was low. Thus, the change of functional groups dominated the effect of electrochemical modification on methane adsorption and desorption behavior in fine coals, while the change of mesopore structures contributed less.
3. APPLICATION OF THE ELECTROCHEMICAL METHOD

The electrochemical method can be used to accelerate methane extraction alone or combined with the hydraulic fracturing method. Hydraulic fracturing technology provides a good electrolyte environment for the electrochemical modification method, which can effectively improve the methane desorption rate and accelerate the discharge of residual water from coal seams and promote gas emission. Studies by Kuh and Kim have shown that electrochemical methods are very effective in removing water from coal.29 Previous studies have shown that uniform distribution of electroosmotic flow and good directional controllability effectively solve the pore sensitivity of seepage.30,31 Therefore, it is theoretically feasible to combine the electrochemical modification method with the hydraulic fracturing technology in CBM extraction engineering.

It may be suggested that electrochemical modification methods may cause pollution to groundwater resources in practical engineering applications. Research and application of electrokinetic remediation technology have solved this problem well. Electrokinetic remediation (ER) technology has been used to remove underground pollutants.31−34 Boulakradeche et al. found that organic pollutants from oil production and petrochemical industry and their impact on soil, groundwater, and the surrounding ecosystem can be removed by electrokinetic remediation.33 So, the ions in the electrolyte and the ions in the minerals can eventually be reacted and removed by the use of ER, which suggests that the electrochemical method is environmentally friendly and is promising for applications.

Table 2. Test Results of the Pore Characteristics in the Anode Zone

| program | surface area (m²/g) | pore volume (cm³/g) |
|---------|---------------------|---------------------|
|         | <10 nm | 10−100 nm | >100 nm | all | <10 nm | 10−100 nm | >100 nm | all |
| T1      | 3.1417 | 0.1510    | 0.0710  | 3.3657 | 0.002010 | 0.008890 | 0.001349 | 0.004249 |
| T2      | 2.6237 | 0.2000    | 0.0680  | 2.8917 | 0.002085 | 0.001177 | 0.001475 | 0.004677 |
| T3      | 3.8287 | 0.1540    | 0.0900  | 4.0727 | 0.002261 | 0.001127 | 0.001888 | 0.005276 |
| T4      | 6.1179 | 0.4380    | 0.1230  | 6.6789 | 0.004243 | 0.002341 | 0.002348 | 0.008932 |

Table 3. Test Results of the Pore Characteristics in the Cathode Zone

| sample | surface area (m²/g) | pore volume (cm³/g) |
|--------|---------------------|---------------------|
|        | <10 nm | 10−100 nm | >100 nm | all | <10 nm | 10−100 nm | >100 nm | all |
| T1      | 3.1417 | 0.1510    | 0.0710  | 3.3657 | 0.002010 | 0.008890 | 0.001349 | 0.004249 |
| T2      | 2.0185 | 0.1920    | 0.0640  | 2.2745 | 0.001714 | 0.001029 | 0.001279 | 0.004022 |
| T3      | 3.4217 | 0.2750    | 0.0820  | 3.7787 | 0.002814 | 0.001530 | 0.001770 | 0.006114 |
| T4      | 2.2503 | 0.2690    | 0.0930  | 2.6123 | 0.001953 | 0.001565 | 0.001942 | 0.005460 |
4. CONCLUSIONS

(1) The electrochemical modification was efficient for accelerating methane production, and higher electric potential gradient (4 V/cm) performs best for decreasing methane adsorption and increasing methane desorption in lean coal via electrochemical modification.

(2) Modified coal samples in the anode zone always adsorbed less methane, and the desorption ratio was higher than that in the cathode zone, which indicates that samples in the anodic zone had a better effect than the cathodic zone.

(3) Higher electric potential gradient performs better in the electrochemical modification, which is due to an increase in acid groups in molecular structure and the change of specific surface area of lean coal after modification.

5. EXPERIMENTS

5.1. Coal Samples. The coal samples were collected in Dongqu Coal Mine of the Shanxi Coking Coal Group. The mean maximum vitrinite reflectance $R_{0,\text{max}}$ (%) of the sample was tested according to GB/T 6948-2008 and ISO 7404-5:1994, GB/T 212-2008, and GB/T1574-2007, respectively. The results are listed in Table 4. The mean maximum vitrinite reflectance was 1.88%, and the test sample was lean coal ($1.8 < R_{0,\text{max}} (%) < 1.9$).

5.2. Experimental Apparatus. A schematic diagram of the self-developed electrochemical modification apparatus is shown in Figure 7. The apparatus is mainly composed of an electrolyzer, a DC power supply, a wire, an ammeter, electrode plates, and potential probes. The electrolyzer is 110 mm long, 60 mm wide, and 80 mm high, which was filled with a 0.05 mol/L Na$_2$SO$_4$ electrolyte. The output voltage range of the DC power supply (Dahua Radio Instrument Factory DH1722A-2, China) is 0–110 V, and the output current range is 0–3 A. The anode and cathode electrode plates are graphite electrode plates with a size of 60 mm long, 100 mm high, and 5 mm thick. The potential copper probe can monitor the potential changes in different areas of the electrolyzer in real time.

5.3. Experimental Process. The coal was treated in four different potential gradients. The sample immersed in Na$_2$SO$_4$ solution without an external electric field was represented as T1. The samples modified in a Na$_2$SO$_4$ electrolyte at potential gradients of 1, 2, and 4 V/cm were represented as T2, T3, and T4, respectively. Before the modification, the coal was crushed, from which the particles with diameters of 180–200 μm were selected and dried to constant weight in a vacuum oven at 373.25 K. Then, 100 g of the dried sample was fully saturated in a Na$_2$SO$_4$ electrolyte and treated with 1, 2, and 4 V/cm external electric field intensities for 120 h.

The adsorption/desorption characteristics of methane on T1, T2, T3, and T4 were determined by the analyzer shown in Figure 8. Before the experiment, the coal sample was degassed at 378.15 K for 300 min under vacuum conditions. Then, the sample was cooled to 293.15 K at room temperature. The adsorption equilibrium experiment was carried out at a pressure of 0–7 MPa. When the adsorption experiment was completed, the desorption characteristics of the sample were determined. The desorption rate of methane decreased with time. When the desorption rate of methane was less than 10 cm$^3$/h, it was considered to reach desorption equilibrium.

Table 4. Test Results of the Lean Coal Sample

| $R_{0,\text{max}}$ (%) | proximate analysis (%) | ultimate analysis (%) |
|----------------------|-----------------------|-----------------------|
| 1.88                 | moisture 0.98          | ash yield 21.29       |
|                      | volatile matter 16.16  | C 90.32               |
|                      |                       | H 4.29                |
|                      |                       | O+S 2.12              |
|                      |                       | N 1.36                |

Figure 7. Schematic of the electrochemical modification device.

Figure 8. Experimental setup used for the adsorption and desorption measurements.
The surface functional groups of T1, T2, T3, and T4 modified were characterized by FTIR (Thermo Fisher Nicolet iS5, USA). The traditional KBr tablet method was used for the FTIR test. Both the samples and KBr were dried in a vacuum oven at 353 K for 12 h to minimize the impact of moisture. Then, 1 mg of coal and 200 mg of KBr were ground and pressed into pellets. The pellets were analyzed at room temperature with a scanning range of 450–4000 cm\(^{-1}\).

Liquid nitrogen adsorption is usually used to characterize the mesopores of coal. Before the determination, all samples were dried in a vacuum oven at 373.15 K. Nitrogen with 99.99% purity was used in the experiment, and the temperature was maintained at 77.35 K.

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**Notes**

The authors declare no competing financial interest.
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