Experimental Study on the Thermal Effect during Gas Adsorption and Desorption on the Coal Surface

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ABSTRACT: The thermal effect of coal adsorption/desorption gas is very important for understanding the evolution of coal temperature and interaction between coal and gas during coal and gas outburst. The pressure difference between the high gas pressure area in front of the working face and the low gas pressure area near the coal wall may affect the adsorption/desorption thermal effect. In order to reveal the characteristics of the coal adsorption/desorption gas thermal effect at different pressure differences, a thermo-hydro-mechanical-coupled experimental system of coal and gas was designed. Taking no.3 coal from Xinjing Mine as the research object, the characteristics of the coal adsorption/desorption gas thermal effect under different pressure differences are studied by using the cycle-step experiment method. It is found that coal adsorbs gas to release heat, while coal desorbs gas to absorb heat. Also, the temperature variation and temperature accumulation caused by adsorption are greater than those caused by desorption. Under the same pressure difference, the temperature increase rate during the adsorption changes from large to small, and the temperature variation gradually decreases; the temperature decrease rate during the desorption changes from small to large, and the temperature variation gradually decreases; the temperature decrease rate caused by adsorption are greater than those caused by desorption. Under the same pressure difference, the temperature increase rate during the adsorption changes from large to small, and the temperature variation gradually decreases; the temperature decrease rate during the desorption changes from small to large, and the temperature variation gradually increases; desorption is the reverse process of adsorption. The relation between temperature variation and gas pressure is linear, and the increasing range of temperature variation gradually decreases with the increase of pressure difference. The relation between temperature accumulation and gas pressure conforms to an exponential function, and the decreasing range of temperature accumulation gradually decreases with the increase of pressure difference. The greater the pressure difference, the greater is the energy variation caused by the adsorption/desorption thermal effect. The experimental results of different pressure differences can reflect the characteristics of the coal adsorption/desorption gas thermal effect under different geological structures or outburst types.

1. INTRODUCTION

Coal and gas outburst is one of the main disasters in coal mines. The process of coal and gas outburst is accompanied by the accumulation and release of energy, and it has a strong destructive power. Strengthening prediction of coal and gas outburst is an important way to ensure mine safety. At present, there are plenty of indexes to prediction of coal and gas outburst at home and abroad, such as drill cuttings gas desorption index, drillings volume index, index of initial velocity of gas emission from borehole, and temperature index. Among them, the fundamental question involved in predicting the risk of outburst using temperature indicators are temperature changes caused by coal deformation, gas adsorption/desorption, and gas seepage. The temperature changes in the process of coal and gas outburst mainly include the heat released by coal deformation and adsorbed gas, the heat absorbed by gas expansion and desorption, and the heat diffusion and convection in the process of gas seepage. The adsorption/desorption thermal effect is an important part of coal temperature change. Temperature rise promotes the desorption–diffusion–seepage of gas, while the thermal expansion strain leads to the decrease of coal permeability and inhibits the desorption–diffusion–seepage of gas. At present, the mechanism of the internal temperature change of coal on the mutual coupling relation between coal and gas is still unclear. Therefore, studying the thermal effect of coal adsorption/desorption gas can provide a theoretical basis for exploring the mutual coupling relation between coal and gas. In addition, different geological structures and outburst types have different pressure differences between the high gas pressure area of coal in front of working face and the low gas pressure area near the coal wall. Also, the pressure difference has an important influence on the thermal effect of coal adsorption/desorption gas.

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work and achieved fruitful results. The analysis of the evolution law and measurement of coal adsorption/desorption gas temperature variation are important contents to study the thermal effect of adsorption/desorption. Jagiello et al. gave a calculation method of the maximum volume of work, which can be performed by a gas while being liberated from a coal bed. Glass and Larsen used inverse gas chromatography to get adsorption heats and entropies. Turrini et al. studied the adsorption and desorption of carbon dioxide and methane on three samples of hard coal with different C_daf contents and on the active carbon. The experimental data were described using the thermal sorption equation of the virial form. Myers set up the thermodynamic equation for the adsorption of multi-component gas mixtures in microporous adsorbents based on the principle of solution thermodynamics. Ibach presented general arguments about the sign of the shift in the heat of adsorption with strain and on the general trends in the coverage dependence of the effect. He and Seaton used the Monte Carlo method to simulate the isosteric heat of adsorption of methane in an activated carbon adsorbent. Sakurovs et al. presented sorption curves of three dry Argonne Premium coals, for carbon dioxide, methane, and nitrogen at two different temperatures at pressures up to 15 MPa. The analysis results of the adsorption curve show that the calculated heat of sorption for these gases on these coals is similar to those found for these gases on activated carbon. Richard et al. proposed that simulations of the thermal effects during adsorption cycles are a valuable tool for the

Figure 1. Law of temperature change during adsorption under different pressure differences; (a) adsorption temperature (0.2 MPa); (b) rate of temperature change (0.2 MPa); (c) adsorption temperature (0.4 MPa); (d) rate of temperature change (0.4 MPa); (e) adsorption temperature (0.6 MPa); and (f) rate of temperature change (0.6 MPa).
design of efficient adsorption-based systems such as gas storage, gas separation, and adsorption-based heat pumps. Simulations of the thermal phenomena associated with hydrogen, nitrogen, and methane adsorption on activated carbon for supercritical temperatures and high pressures was carried out. Rahman et al.\textsuperscript{23,24} reported the theoretical frameworks for the thermodynamic quantities, namely, the heat of adsorption, specific heat capacity, entropy, and enthalpy for the adsorption of methane onto various carbonaceous materials. Temperature variation causes deformation, and deformation generates heat, which causes temperature variation. Based on the molecular migration and energy conversion during the adsorption/desorption, the thermal deformation effect of adsorption/desorption is proposed.\textsuperscript{25} Askalany et al.\textsuperscript{26} proposed a thermodynamic formalism on adsorbed phase volume which is a function of adsorption pressure and temperature for the precise estimation of the isosteric heat of adsorption. Based on the study of surface temperature distribution characteristics of anthracite samples during the gas adsorption/desorption, a new method for calculating adsorbed quantity of gas in coal based on temperature increment is provided.\textsuperscript{27} The isothermal gas adsorption experiments of coal with different initial free gas pressures at different temperatures were conducted, and a mathematical function of the adsorption heat was established.\textsuperscript{28} By using the coal gas adsorption gas temperature experiment system, the evolution law of coal temperature and the influence of adsorption equilibrium pressure and surrounding temperature on the temperature change of coal were studied.\textsuperscript{29}

Figure 2. Law of temperature change during desorption under different pressure differences; (a) desorption temperature (0.2 MPa); (b) rate of temperature change (0.2 MPa); (c) desorption temperature (0.4 MPa); (d) rate of temperature change (0.4 MPa); (e) desorption temperature (0.6 MPa); and (f) rate of temperature change (0.6 MPa).
In summary, scholars have studied the influence of temperature field on the mutual coupling relation between coal and gas by changing the external temperature environment. They have not studied the coupling relation between seepage field and stress field under the action of temperature changes inside coal. The existing research ignores the influence of ground stress on coal samples, the experimental method consists mostly of constant pressure adsorption/desorption, and the quantitative research results are relatively few. In addition, there is a lack of research on the characteristics of coal adsorption/desorption gas thermal effects under different pressure differences. In order to explain the law of temperature change caused by coal adsorption and desorption, using the self-designed coal and gas thermo-hydro-mechanical-coupled experiment system and the cyclic-step adsorption/desorption experiment method, the influence of the gas pressure difference on the adsorption/desorption thermal effect of the coal sample under loading conditions was carried out in a constant temperature environment. It provides a theoretical basis for studying the mutual coupling relation between coal and gas and using temperature indicators to predict the risk of coal and gas outbursts.

2. RESULTS

2.1. Temperature Variation during the Process of Gas Adsorption. As shown in Figure 1, when the pressure differences are 0.2 and 0.4 MPa, the temperature variation of the coal sample gradually decreases, the time for the coal sample temperature to reach the extreme value is prolonged, and the temperature change rate is changed from large to small; the temperature evolution curve with a pressure difference of 0.6 MPa shows that after the gas pressure is increased by 0.6 MPa on the basis of the initial inflation pressure, the temperature variation is greater than the temperature variation under the initial inflation pressure. The main reason is that the pressure difference is greater than the initial inflation pressure. As the pressure increases to 1.6 MPa, the evolution law of coal sample temperature is the same as that when the pressure differences are 0.2 and 0.4 MPa.

The temperature variation gradually decreases in the process of the adsorption at the same pressure difference. When the pressure difference is 0.2 MPa, the temperature variation is reduced from 0.5 to 0.06 °C and the time for the coal sample temperature to reach the maximum value is extended from 3565 to 6876 s; when the pressure difference is 0.4 MPa, the temperature variation is reduced from 0.5 to 0.32 °C and the time for the coal sample temperature to reach the maximum value is extended from 3540 to 6094 s; and when the pressure difference is 0.6 MPa, the coal sample temperature variation is reduced from 0.5 to 0.41 °C and the time for the coal sample temperature to reach the maximum value is extended from 3572 to 4946 s. The temperature change rate increases with the increase of the gas pressure difference.

2.2. Temperature Variation during the Process of Gas Desorption. As shown in Figure 2, when the pressure differences are 0.2 and 0.4 MPa, the temperature variation of the coal sample gradually increases, the time for the coal sample temperature to reach the extreme value is shortened, and the temperature change rate is changed from small to large; the temperature evolution curve with a pressure difference of 0.6 MPa shows that after the gas pressure is reduced by 0.6 MPa for the second time, the temperature variation is greater than the temperature variation caused by the third experiment. The main reason is that the pressure difference is greater than the remaining gas pressure in the system during the third experiment. The evolution law of the coal sample temperature is the same as that when the pressure differences are 0.2 and 0.4 MPa.

The temperature variation gradually increases in the process of desorption at the same pressure difference. When the pressure difference is 0.2 MPa, the temperature variation is increased from 0.06 to 0.48 °C and the time for the coal sample temperature to reach the minimum value is shortened from 6124 to 3190 s; when the pressure difference is 0.4 MPa, the temperature variation is increased from 0.31 to 0.48 °C and the time for the coal sample temperature to reach the minimum value is shortened from 4486 to 3281 s; and when the pressure difference is 0.6 MPa, the coal sample temperature variation is increased from 0.39 to 0.48 °C and the time for the coal sample temperature to reach the minimum value is shortened from 4283 to 2990 s. The desorption experiment is the reverse process of the adsorption experiment.

3. DISCUSSION

3.1. Relation between Temperature Variation and Pressure Difference. Figure 3 shows the adsorption quantity and temperature variation under different pressure differences. Comparing the change trend of the adsorption quantity and
the temperature variation, we can see that the larger the adsorption quantity, the more is the heat released during the adsorption and the faster is the temperature rise of coal. The temperature variation has a linear relation with the gas pressure. When the pressure difference is constant, the adsorption quantity gradually decreases with the increase of gas pressure. When the adsorption is close to saturation, the gas pressure variation decreases, and the adsorption rate is close to zero. At the same time, the adsorption quantity decreases rapidly, and the heat released during the adsorption reduces, which causes a decrease in the temperature variation of the coal sample. As the pressure difference increases, the slope of the temperature variation curve gradually decreases. The increase of the pressure difference is equivalent to the increase of gas pressure, which causes a rapid increase in temperature variation. When the desorption experiment approaches the equilibrium state, the desorption rate is close to zero. Therefore, the desorption quantity decreases rapidly, and the temperature variation in the coal surface decreases. As the pressure difference increases, the slope of the temperature variation curve gradually increases. The increase in the pressure difference indicates that the amount of gas pressure drop in the desorption experiment increases, which causes the increase of desorption quantity. It leads to an increase in the amount of temperature drop of the coal sample surface.

Comparing the change trend of the desorption quantity and the temperature variation, we can see that the larger the desorption quantity, the more is the heat absorbed during the desorption and the faster is the temperature drop of coal. The temperature variation has a linear relation with the gas pressure. When the pressure difference is constant, the desorption quantity gradually increases with the decrease of gas pressure, which causes a rapid increase in temperature variation. When the desorption experiment approaches the equilibrium state, the desorption rate is close to zero. Therefore, the desorption quantity decreases rapidly, the heat absorbed during the desorption reduces, and the temperature variation in the coal surface decreases. As the pressure difference increases, the slope of the temperature variation curve gradually increases. The increase in the pressure difference indicates that the amount of gas pressure drop in the desorption experiment increases, which causes the increase of desorption quantity. It leads to an increase in the amount of temperature drop of the coal sample surface.

In order to deeply analyze the relation between the pressure difference and the temperature variation, the relation between the temperature variation and the gas pressure in the process of adsorption/desorption under different pressure differences was fitted (Figures 3b and 4b). In the adsorption experiment, when the gas pressure is 1.6 MPa and the pressure difference increases from 0.2 to 0.4 MPa, the temperature variation increases by 0.25°C; when the pressure difference increases...
from 0.4 to 0.6 MPa, the temperature variation increases by 0.1 °C. In the desorption experiment, when the gas pressure is 1.6 MPa and the pressure difference increases from 0.2 to 0.4 MPa, the temperature variation increases by 0.24 °C; when the pressure difference increases from 0.4 to 0.6 MPa, the temperature variation increases by 0.09 °C. Under the same pressure difference, the temperature variation caused by adsorption is greater than the temperature variation caused by desorption, and the time for the temperature to reach the maximum value during the adsorption is longer than the time for the temperature to reach the minimum value during the desorption. Adsorption expansion strain and pore compression strain cause the coal sample to deform, which changes the characteristics of coal adsorbing gas. Therefore, the adsorbed gas cannot be completely transformed into a free state.

According to the experimental method reported in this article, the adsorption quantity of the coal sample is less than the adsorption quantity and the heat released by adsorption is greater than the heat absorbed by desorption. The heat absorbed or released by the coal sample during the adsorption/desorption when the pressure difference increases from 0.2 to 0.4 MPa is greater than those when the pressure difference increases from 0.4 to 0.6 MPa. As the pressure difference increases, the increasing range in temperature variation gradually decreases. According to the Langmuir adsorption theory, there is a limit value for the adsorption quantity of coal. The increasing range in the variation of adsorption quantity gradually decreases when the pressure difference increases. As a result, the increasing range in temperature variation of the coal sample gradually decreases.

3.2. Relation between Temperature Accumulation and Pressure Difference. Figure 5 shows the cumulative adsorption quantity and cumulative desorption quantity under different pressure differences. Comparing the law of cumulative adsorption quantity and cumulative desorption quantity change, it can be seen that under the experimental conditions of cycle-step adsorption/desorption, the cumulative adsorption quantity is greater than the cumulative desorption quantity. Cumulative adsorption quantity and cumulative desorption quantity have an exponential function relation with gas pressure. When the pressure difference is constant, the difference between the cumulative adsorption quantity and the cumulative desorption quantity gradually increases. As the pressure difference increases, the difference between the cumulative adsorption quantity and the cumulative desorption quantity under the same pressure difference gradually decreases. Comparing Figures 5 and 6, it can be seen that the change trend of cumulative adsorption quantity and cumulative desorption quantity is similar to that of temperature accumulation.

In order to further analyze the relation between the pressure difference and the temperature accumulation, the relation between the temperature accumulation and the gas pressure in the adsorption/desorption under different pressure differences was fitted (Figure 6). The fitting equation is shown in Table 1. It can be seen from the figure that the temperature accumulation during the adsorption/desorption has an exponential function relation with the gas pressure. As the pressure difference increases, the temperature accumulation gradually decreases. When the pressure difference is constant, the temperature accumulation during the adsorption is greater than that during the desorption. The temperature accumulation variation when the pressure difference increases from 0.2 to 0.4 MPa is greater than that when the pressure difference increases from 0.4 to 0.6 MPa. The decreasing range of the temperature accumulation gradually decreases with the increase of the pressure difference.

The existing research results show that the amount of deformation caused by stepwise loading and constant pressure loading is different, and the amount of deformation caused by stepwise loading is greater than that caused by constant pressure loading. Therefore, the deformation of the coal sample in the adsorption decreases with the increase of the pressure difference, the increasing range in the adsorption quantity of the coal sample decreases, and the increasing range in the temperature accumulation of the coal sample surface decreases. Desorption is the reverse process of adsorption, and the adsorption/desorption experiment is carried out cyclically. Therefore, the change law of the temperature variation and temperature accumulation in the initial stage of desorption is consistent with the law of that in the later stage of adsorption.32,33 Under the same pressure difference, the coal sample temperature variation and temperature accumulation during the adsorption are greater than those during the desorption. The adsorption expansion deformation of coal is greater than the desorption shrinkage deformation during the adsorption/desorption desorption. Therefore, the adsorption/desorption process of coal is not completely reversible, and the adsorbed gas cannot be completely transformed into a free state.

In summary, the greater the pressure difference, the greater is the energy variation caused by the adsorption/desorption thermal effect and the smaller is the energy accumulation. The adsorption/desorption thermal effect directly changes the equilibrium state of free gas and adsorbed gas in coal and the gas seepage velocity. In addition, the thermal expansion strain caused by the adsorption/desorption thermal effect causes the effective stress to change, which affects the permeability characteristics of coal. Therefore, the adsorption/desorption thermal effect is an important factor affecting
the mutual coupling relation between coal and gas. The experimental results of different pressure differences correspond to the characteristics of the adsorption/desorption thermal effect under different geological structures or outburst types.

4. CONCLUSIONS

Gas pressure difference has a great influence on the adsorption/desorption thermal effect. This paper analyzes in detail the characteristics of the adsorption/desorption thermal effect under different pressure differences and establishes the relation between temperature variation and gas pressure and the relation between temperature accumulation and gas pressure. The research results provide a theoretical basis for predicting the risk of coal and gas outburst using temperature indicators and improve the mutual coupling relation between coal and gas.

(1) Under the same pressure difference, the evolution law of the coal sample temperature during the adsorption is that the rate of temperature rise changes from large to small, the temperature variation gradually decreases, and the time for the temperature to reach the maximum value is prolonged. The evolution law of the coal sample temperature during the desorption is that the rate of temperature drop changes from small to large, the temperature variation gradually increases, and the time for the temperature to reach the maximum value is shortened.

(2) The adsorption of gas is an exothermic process, while the desorption of gas is an endothermic process. Under the same pressure difference, the temperature variation during the adsorption is greater than that during the desorption; the temperature accumulation during the adsorption is greater than that during the desorption.

(3) The relation between the temperature variation and the gas pressure is linear, and the increasing range of the temperature variation gradually decreases with the increase of the pressure difference. The relation between the temperature accumulation and the gas pressure conforms to an exponential function, and the decreasing range of temperature accumulation gradually decreases with the increasing pressure difference.

(4) The change law of adsorption/desorption quantity is similar to that of temperature variation. The change law of cumulative adsorption/desorption quantity is similar to that of temperature accumulation.

(5) The greater the pressure difference, the greater is the energy variation caused by the adsorption/desorption thermal effect. The adsorption/desorption thermal effect changes the equilibrium state of free gas and adsorbed gas in coal, the gas seepage velocity, and permeability characteristics of coal. The adsorption/desorption thermal effect is an important factor affecting the mutual coupling relation between coal and gas.

5. EXPERIMENTAL SECTION

5.1. Experimental System. Using the self-designed true triaxial coal adsorption/desorption gas experiment device, the experiment of temperature change during coal adsorption/desorption under constant temperature and loading conditions was completed. The experiment system mainly includes pore pressure loading and unloading device, true triaxial adsorption/desorption device, stress loading device, vacuuming device, thermostatic device, drainage and gas collecting device, and data acquisition system (Figure 7). Temperature data acquisition is achieved by attaching a high-precision chip platinum resistance temperature sensor to the surface of the coal sample.

5.2. Coal Samples and Experimental Methods. The coal sample used in the experiment was taken from Xinjing coal mine of Yangmei group, which belongs to the high gas outburst mine. The main mining no. 3 coal seam is an outburst coal seam. The raw coal sample is obtained at the driving working face. All coal samples were packaged tightly at the scene and transported to the laboratory in time. The coal sample is cut into 5 cm × 5 cm × 10 cm square specimen, and then it is polished and leveled by grinding machine; the flatness of the end face is ±1 mm. Figure 8 shows the prepared experiment coal sample, and Table 2 shows the basic parameters of the coal sample.

To study the temperature evolution law of coal adsorption/desorption at different gas pressure differences, the experiment of coal cycle-step adsorption/desorption was designed. The experiment methods are shown in Table 3. The initial inflation pressure is 0.4 MPa, and the gas pressure differences are 0.2, 0.4, and 0.6 MPa/time. The initial pressure of the desorption experiment is the maximum adsorption equilibrium pressure of the adsorption experiment, and the ambient temperature is maintained at 30 °C.
5.3. Experimental Procedures. This experiment measured the temperature change of raw coal during the process of gas adsorption/desorption under different pressure differences. The initial gas pressure is the same, and the pressure is gradually increased to the target pressure with a certain gas pressure difference. Then, it is gradually reduced to the minimum pressure with the same gas pressure difference.

Step 1: Check the airtightness of the device and then install the coal sample. Connect the temperature measurement system and the gas pressure measurement system. Open the incubator and debug the oven temperature to 30°C. Turn on the data acquisition system.

Step 2: Turn on the triaxial stress loading system and adjust the axial pressure and the confining pressure to a predetermined value.

Step 3: Connect the high pressure gas cylinder to the experiment system.

Step 4: Turn on the vacuum pump to vacuum degas the gas pressure loading system.

Step 5: Conduct an experiment of coal cycle-step adsorption gas. Adjust the pressure relief valve at the outlet of the high pressure gas cylinder to the required pressure value and ensure that the gas pressure in the standard tank reaches the predetermined value. After the gas pressure in the standard tank is stabilized, the gas in the standard tank is introduced into the adsorption/desorption device, and the adsorption process lasts for 12 h. Besides, the temperature variation and gas pressure during the gas adsorption must be recorded.

Step 6: Conduct an experiment of coal cycle-step desorption gas. After the adsorption experiment is cycled to the target pressure, the gas pressure in the adsorption/desorption device is reduced by an equal pressure difference, and the desorption process lasts for 12 h. The record of the experimental data is the same as the step 6.

Step 7: Repeat step 5 and step 6 and adjust the gas pressure difference to 0.2, 0.4, and 0.6 MPa/time.

Table 2. Basic Parameters of the Coal Sample

| coal       | technical analysis | adsorption constant | gas pressure |
|------------|--------------------|---------------------|--------------|
|            | M_{daf}/%          | A_{ddf}/%           | V_{daf}/%     | a/m³·t⁻¹ | b/MPa⁻¹ | /MPa |
| anthracite | 1.19               | 6.31                | 8.25         | 34.76    | 1.5335  | 1.3–2.6 |

Table 3. Experimental Method

| pressure difference (MPa/time) | gas pressure loading path (MPa) |
|--------------------------------|---------------------------------|
| 0.2                            | 0.4–0.6–0.8–1.0–1.2–1.4–1.6    |
| 0.4                            | 0.4–0.8–1.2–1.6–1.2–0.8–0.4    |
| 0.6                            | 0.4–1.0–1.6–1.0–0.4            |

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