

Rapid Contrastive Experimental Study on the Adiabatic Spontaneous Combustion Period of Loose Lignite

Ziwen Dong,* Lifeng Sun, Tinggui Jia, Shengli Guo, Weile Geng, Wenhui Yu, Zikun Pi, Guangli Li, and Chen Luo



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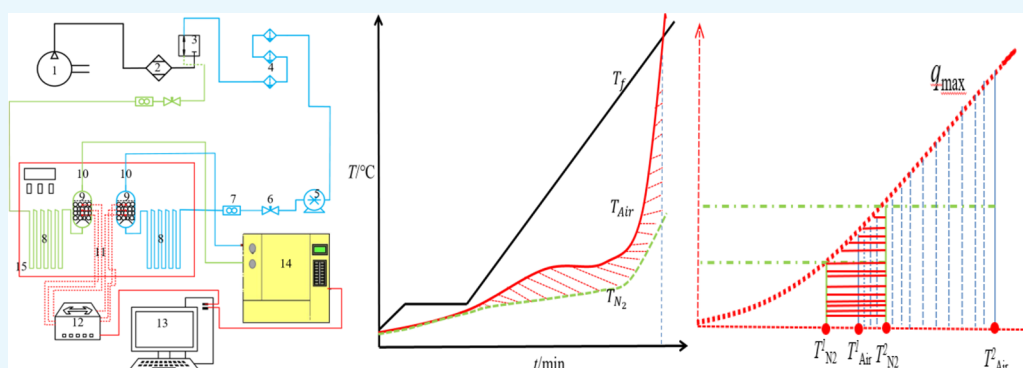


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ABSTRACT: The adiabatic spontaneous combustion period of coal is an important index for the macroscopic characterization of coal spontaneous combustion, and it is affected by many internal and external factors. There are several methods to study it, but there are various shortcomings to these methods. Some require too much time, while others have too many interfering factors. To quickly obtain the accurate adiabatic spontaneous combustion period of coal, a rapid contrastive experimental method was designed. In this method, the coal samples of the experimental and control groups were the same, and air and nitrogen were used as control atmospheres. A theoretical calculation method for the adiabatic spontaneous combustion period based on this method is proposed. The experimental results showed that during the temperature-programmed coal spontaneous combustion experiment, the increase in the coal temperature was due to physical and chemical heating. Physical heating is the heating effect of the temperature-programmed furnace body and the heated gas on the coal sample. Chemical heating includes oxidative exothermic heating promoted by physical and adiabatic oxidation heating. The adiabatic oxidative heat release can be determined by the oxidation heat release in the air atmosphere minus the oxidation heat release corresponding to the coal sample temperature in the nitrogen environment at the same period. The adiabatic spontaneous combustion period of coal can be calculated from the adiabatic oxidation heat release. Our results provide a rapid contrastive experimental method to quickly obtain the accurate adiabatic spontaneous combustion period of coal.

1. INTRODUCTION

The spontaneous combustion of coal is a well-known phenomenon worldwide.¹ It significantly affects and restricts the safe production of coal mines, threatens the lives and health of miners, and causes greenhouse gas emissions and environmental pollution.^{2–4} Moreover, it significantly affects other coal-related activities, such as the storage, transportation, processing, and utilization of coal.⁵ After coal contacts oxygen, oxidation reactions occur. Under the comprehensive influence of internal and external factors, when heat accumulates, the temperature of the coal increases continuously, and spontaneous combustion occurs. There are many mathematical models and experimental and statistical methods to study the spontaneous combustion liability of coal, but no specific

method has become the standard to predict the spontaneous combustion liability.^{6–8}

Various experimental methods have been developed, including crossing point temperature (XPT) analysis, gas index analysis, Russian U-index analysis, Olpinski index analysis, adiabatic calorimetry, self-heating temperature analysis, peroxy complex analysis, differential thermal analysis (DTA), the Stage II Slope method, wet oxidation potential

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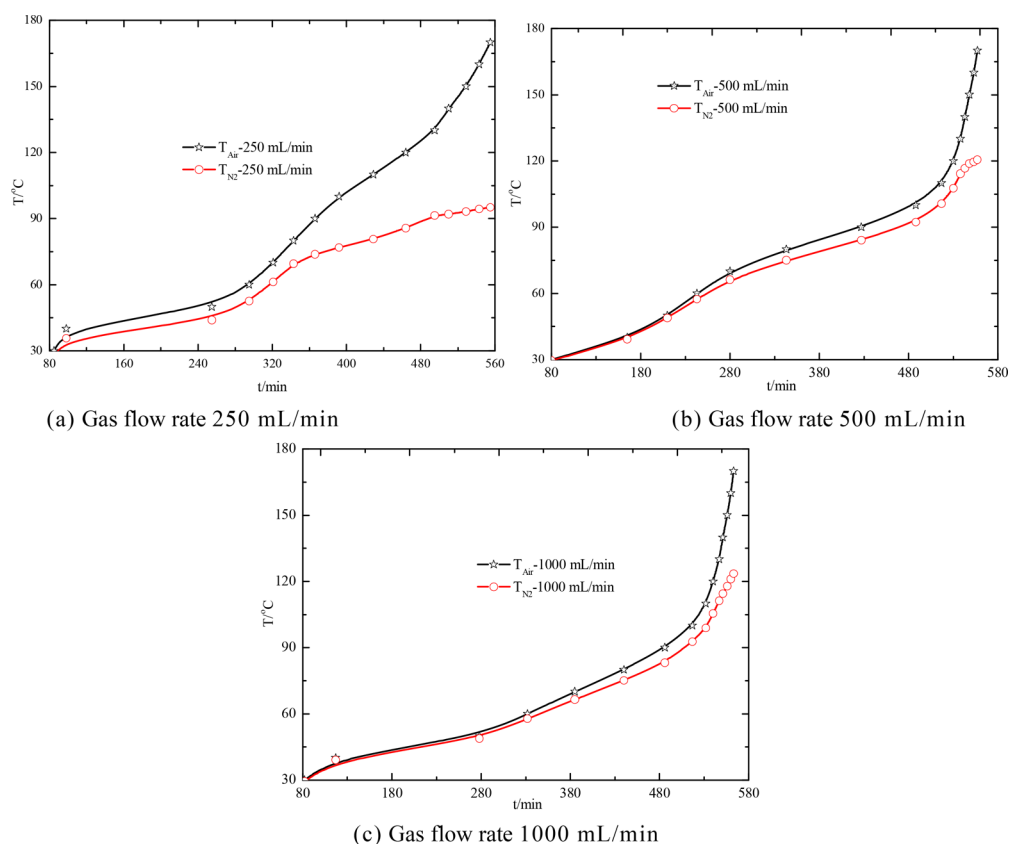


Figure 1. Temperature of coal samples varies with time under different air and nitrogen gas flow rates.

(WOP) analysis, modified XPT index analysis, FCC (Feng, Chakravorty, Cochrane) analysis, and Wits-Ehac index analysis.^{6–10} To evaluate the spontaneous combustion liability of coal, Gouws et al.¹¹ developed an adiabatic calorimetry method, and Zhang et al.¹² developed a nested spontaneous combustion furnace with better thermal insulation performances.

The spontaneous combustion of coal is a gradual process that requires a certain amount of time.¹³ Cheng et al.¹⁴ and Beamish et al.¹⁵ established large and small experimental test systems, respectively, to study coal spontaneous combustion by fully simulating the heating process of coal spontaneous combustion. Based on field studies, Xu et al.¹⁶ and Wen et al.¹⁷ have built several large experimental platforms of different scales to study coal spontaneous combustion and simulate the process of coal spontaneous combustion on site. Deng et al.¹⁸ conducted a large number of low-temperature oxidation and heating experiments using XK (Xi'an University of Science and Technology) series coal spontaneous combustion experimental platforms. Li et al.¹⁹ analyzed the variations of the high-temperature point and the characteristic gas using a large-scale experimental platform for coal spontaneous combustion and accurately tested the shortest coal spontaneous combustion period.

Lu et al.²⁰ improved adiabatic experimental equipment based on the basic equations of coal spontaneous combustion dynamics and proposed the self-activation reaction theory in the process of coal spontaneous combustion by analyzing experimental data. Tan et al.,²¹ Zhu et al.,²² and Wang et al.^{23,24} also established adiabatic heating systems, studied the influence of the oxygen concentration on the adiabatic oxidation characteristics, analyzed the characteristic temper-

ature in the adiabatic spontaneous combustion process in detail, and studied the influence of the metamorphosis degree on the characteristic parameters of coal spontaneous combustion using this system. Xu et al.²⁵ designed and built an adiabatic coal spontaneous combustion experimental system.

To eliminate the long adiabatic oxidation test times, Zhong et al.²⁶ proposed a test method of the critical temperature of spontaneous combustion of coal based on temperature-programmed conditions. Huang et al.²⁷ proposed the concept of temperature difference leading and an experimental test method of the adiabatic ignition period based on temperature rise leading. Liang^{28,29} proposed an equal heat flow heating method to heat up the coal sample and study the characteristics of the spontaneous combustion of coal.

Based on the positive temperature difference leading method, Liu³⁰ selected the optimal reference sample ratio of 1:7:2 for corundum, fly ash, and cement by optimizing the heat-transfer properties of reference experimental samples to optimize the design of the heating furnace and achieve precision analysis of the experimental results. A method was proposed to determine the adiabatic oxidation heating behaviors of coal by comparing the temperature of the reference sample with those of coal samples. The method had high accuracy, theoretical feasibility, and reproducibility, and the adiabatic oxidation heat effect and spontaneous combustion period of coal could be quickly measured by using a single leading temperature.

The coal spontaneous combustion period is defined as the time from when the coal body is first exposed to air, begins to undergo natural oxidation, and releases heat to the time when spontaneous combustion occurs. The coal spontaneous

combustion period results from its own spontaneous combustion characteristics and external environmental factors. Even if the thermal properties of coal can be reproduced by using noncoal samples as reference samples, the reference samples cannot reflect the pore structure characteristics of coal. As is well-known, coal has a complex amorphous structure, and the amorphous structure is significantly affected by water immersion or loss. The change of the structure will significantly change the spontaneous combustion characteristics of coal.^{31–33} With the decrease in the particle size, the moisture content, the heat of wetting release, and the risk of spontaneous combustion increase.³⁴ Therefore, it is insufficient to use noncoal reference samples in the experiments of low-rank coal, especially for high-moisture-content coal.^{35,36}

The existing experimental methods can reflect the process of coal spontaneous combustion to a certain extent and have a higher theoretical value, but they also have several shortcomings. The experimental process of a large-scale test bench is more complex and yields low precision and poor repeatability. The experimental periods of adiabatic oxidation experiments are too long, and it is difficult to guarantee the adiabatic environment in the experimental process. Temperature leading experiments can avoid these problems to a certain extent. Furthermore, they have made great progress in the selection of reference samples, and the experimental results have higher accuracy. However, the reference samples cannot accurately reflect the effect of the coal pore structure and moisture on the spontaneous combustion.

Hence, based on previous results, in this study, the samples of the experimental and control groups were from the same coal sample. Both underwent the same process in heating furnace experiments, in which nitrogen was pumped into the control group sample tank and air was pumped into the experimental group sample tank. During the experiments, the temperature of the coal sample was constantly monitored, and the type and concentration of the exhaust gas in the experimental group were analyzed. The oxygen consumption rates, gas production rates, and upper limit oxidation heat release intensities of the coal samples were calculated. Additionally, a method for calculating the adiabatic spontaneous combustion period of coal based on the comparison of the experimental results in nitrogen and air was proposed, and the adiabatic spontaneous combustion periods of coal under different air flow rates were calculated. The results of this study can provide a theoretical reference for the experimental study of spontaneous combustion characteristics and the prevention and control of the spontaneous combustion of coal.

2. EXPERIMENTAL RESULTS AND DISCUSSION

2.1. Analysis of Temperature and Gas Concentration.

With the extension of the heating time, the coal temperature gradually increased, but the coal temperature in the nitrogen atmosphere was lower than that in the air atmosphere. With the extension of the time and the increase in the temperature, the difference between the two temperatures gradually increased, as shown in Figure 4.

The coal heating curves in the nitrogen atmosphere shown in Figure 1 show that under the same heating rate and time, the coal temperatures were different due to the different nitrogen flow rates. The larger the flow rate was, the higher the coal temperature became, indicating that the programmed-temperature furnace and the heated gas had different influences on the physical heating of the coal samples.

Therefore, when studying the characteristics of the spontaneous combustion of coal through temperature-programmed experiments, the heating curves of the coal samples will be distorted if the physical heating effects of the furnace body and the incoming gas are ignored. In this case, the analysis of the coal temperature variations will not be highly representative. Thus, the conventional temperature-programmed experiment can only be used to analyze the coal sample oxidation and gas generation behaviors at the corresponding temperature.

Under the temperature-programmed conditions, the high coal temperature rise in the nitrogen environment belongs to physical heating, the temperature of the coal sample is related to the furnace temperature, nitrogen temperature, nitrogen flow rate, and heating time, and the nitrogen temperature is determined by the furnace temperature, so for the same heat-up time, the furnace temperature and nitrogen flow rate corresponding to different flow rates of nitrogen are also the same: the difference is only found in the nitrogen flow rate, and the effect of the nitrogen flow rate alone is not as significant as that of multiple factors. Therefore, the temperature difference in the paper is not significant, especially in the case of no significant difference in nitrogen flow rate, for example, at the nitrogen flow rates of 500 mL/min and 1000 mL/min in Figure 1b and c.

In an air atmosphere, coal reacts with oxygen via oxidation reactions, producing and releasing CO, CO₂, and some other gases. Because the purpose of this experimental study was to examine the adiabatic natural ignition period of coal, the heat release intensity of coal needs to be calculated. Therefore, we only analyzed the volume concentration changes of O₂, CO, and CO₂. In the temperature-programmed experiments of coal, the air flow rates, oxygen consumption rates, and residual oxygen concentrations were varied. Figure 2 shows the oxygen concentration changes within the range of 30–170 °C in the temperature-programmed process under different air flow rates.

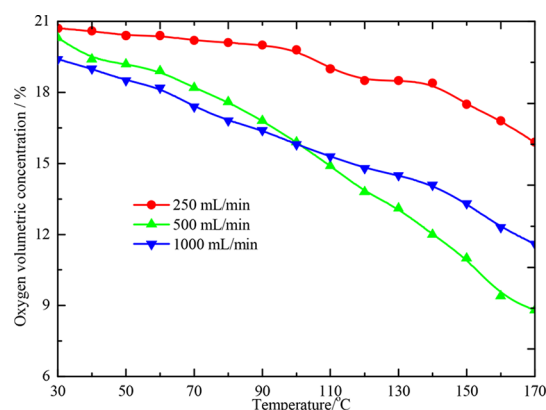


Figure 2. Volume concentration of O₂.

As shown in Figure 2, the oxygen concentration gradually decreased with the increase in the temperature. Within the temperature range of 40–100 °C, the oxygen concentration was the highest at an air flow rate of 250 mL/min, followed by 500 mL/min, and it was the lowest at 1000 mL/min. Within the temperature range of 100–170 °C, the oxygen concentration was the highest at an air flow rate of 250 mL/min.

min, followed by 1000 mL/min, and it was the lowest at 500 mL/min.

With the rise of the coal temperature, the oxygen consumption of coal increases and the oxygen concentration decreases gradually. Under the condition of the same coal quality, the greater the air flow, the more sufficient the oxygen, the more intense the oxidation reaction, the greater the oxygen consumption, and the lower the remaining oxygen concentration. As shown in Figure 2, the oxygen concentration intersects between the air flow rates 500 mL/min and 1000 mL/min, at the coal temperature 100 °C. The reason for the intersection is that the oxygen concentration decreases faster under the air flow rate 500 mL/min than the air flow rate 1000 mL/min. However, the absolute oxygen consumed in the process of coal temperature rising from 30 to 100 °C is the accumulation of the oxygen concentration difference and the total flow difference. Although the oxygen concentration of the two is the same and the concentration difference is similar, the flow rate 1000 mL/min is twice that of 500 mL/min, so the absolute oxygen consumed under the flow rate 1000 mL/min is still higher. So, it is meaningless to compare the variation of oxygen concentration separately; only the rate of oxygen consumption is comparable. The strength of coal's oxygen consumption capacity under different air flow rates should be determined by oxygen concentration combined with air volume and the coal sample's accumulation structure parameters. The oxygen concentration change is only one of the basic parameters for analyzing and calculating the oxygen consumption rate. To compare the strength of coal's reaction capacity with oxygen, the oxygen consumption rate of coal must be solved and analyzed, as shown in Figure 5.

The CO concentration in the sample temperature range 30 °C–90 °C had no significant differences due to the differences in the air flow rates, and the concentration was low. When the coal temperature was higher than 90 °C–100 °C, the CO concentration increased significantly with the increase in the coal temperature, and the CO concentration varied significantly at different air flow rates, as shown in Figure 3. The CO₂

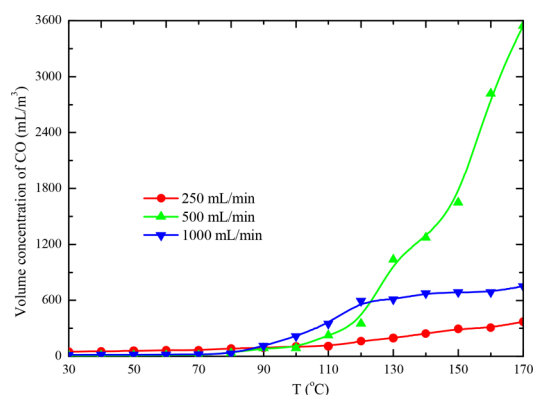


Figure 3. Volume concentration of CO.

concentration varied at different air flow rates. Generally, the CO₂ concentration did not change significantly with the increase in temperature in the range 30–80 °C. When the coal temperature was higher than 80 °C, the CO₂ concentration increased significantly with the increase in the coal temperature, as shown in Figure 4.

The change rule of CO and CO₂ concentration indicates that CO and CO₂ will constantly be produced and the

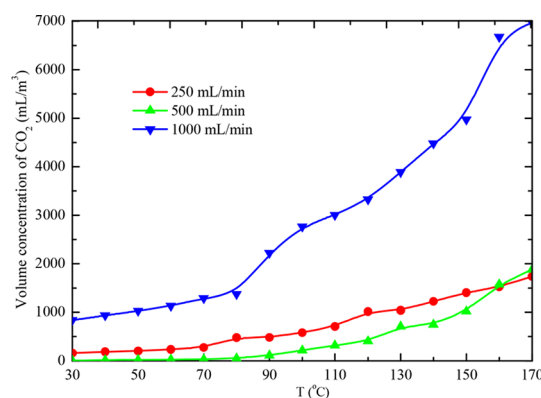


Figure 4. Volume concentration of CO₂.

concentration will gradually increase in the process of coal temperature rising. For the lignite used in this experiment, as shown in Figure 3, when the coal temperature is lower than 90 °C, the concentration of CO and CO₂ is very low. When the coal temperature is higher than 90 °C, the concentration of CO and CO₂ begins to increase rapidly. As shown in Figure 3, when the coal temperature is 90–120 °C, the higher the air volume is, the higher the CO concentration is. When the coal temperature is higher than 120 °C, the CO concentration is the lowest at the air flow rate 250 mL/min, the CO concentration is in the middle at the air flow rate 1000 mL/min, and the CO concentration is the highest at the air flow rate 500 mL/min. Under the condition of 250 mL/min air flow rate, the oxygen supply is insufficient, and the oxidation capacity of coal is weak. Although the oxidation capacity of coal is weak and the oxidation reaction is not sufficient, a lot of CO will be produced, but the air flow rate is 250 mL/min, and the total amount of oxygen that can participate in the oxidation reaction is low, so the amount of CO produced is low. Under this condition, the concentration of CO₂ is also low. This rule can be obtained by comparing Figures 3 and 4. Under the condition of 500 mL/min air flow rate, due to insufficient oxygen supply and incomplete oxidation reaction, the oxygen quantity is significantly higher than that under the condition of 250 mL/min, so the amount of CO produced is relatively large and the concentration of CO₂ is low; this trend can be found through comparative analysis of Figures 3 and 4. Under the condition that the air flow rate is 1000 mL/min, due to sufficient oxygen supply, the oxidation reaction between coal and oxygen is more thorough, so the amount of CO produced by the oxidation reaction is less, and the amount of CO₂ produced will be more, as shown in Figure 4. Similar to the change rule of oxygen concentration, there is no significant comparability in the analysis of the CO and CO₂ concentration alone, so the influence of the air flow rate needs to be analyzed according to the generation rate of CO and CO₂, as shown in Figures 6 and 7.

2.2. Analysis of Gas Reaction and Generation Rate.

During the experiment, the change of the oxygen concentration at each point in the coal sample was mainly related to convection (air flow), diffusion (molecular diffusion and turbulent diffusion), and oxygen consumption through coal–oxygen interactions and other factors. Therefore, the convection–diffusion equation of the oxygen concentration distribution in a coal sample is as follows:

$$\frac{\partial C}{\partial t} = \text{div}(D \cdot \text{grad}(C)) - \text{div}(uC) + V_{O_2}(T)/n \quad (1)$$

where C is the volume concentration of gas in the coal sample (mol/mL), t is time (s), D is the diffusion coefficient of oxygen in the coal sample (m^2/s), u is the average velocity of the air flow in the interspaces (cm/s), $V_{O_2}(T)$ is the oxygen consumption rate (mol/ m^3/s), and n is the porosity.

Under the experimental conditions, because the copper coal sample tank was used and all the temperature sensor inlets and air inlets were sealed, there was no air leakage, and the air mainly flowed along the central axis of the coal sample. Therefore, only the oxygen concentration distribution along the axial direction of the coal sample needed to be considered, and eq 1 becomes the following:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial Z} \left[D \left(\frac{\partial C}{\partial Z} \right) \right] - \frac{\partial uC}{\partial Z} + V_{O_2}(T)/n \quad (2)$$

where Z is the height along the central axis of the coal sample (cm), which is calculated from the gas inlet.

It was assumed that the air only flowed axially and the velocity was constant. Furthermore, the diffusion of oxygen in the experimental coal body and the variation rate of the oxygen concentration with time were neglected, and the coal temperature in the microunit was assumed to be uniform. The oxygen consumption rate can be calculated as follows:

$$V_{O_2}(T)/n = u \frac{dC}{dZ} \quad (3)$$

According to the theory of chemical kinetics and equilibrium, the following equation can be obtained:

$$V_{O_2}(T)/n = K \cdot C \quad (4)$$

According to Arrhenius' law, the oxygen consumption rate is proportional to the concentration of oxygen. Therefore, the oxygen consumption rate in fresh air is expressed as follows:

$$V_{O_2}^0(T) = \frac{C_0}{C} V_{O_2}(T) \quad (5)$$

where C_0 is the concentration of oxygen in fresh air (mol/mL).

The oxygen consumption between Z_1 and Z_2 at any two points on the central axis is calculated as follows:

$$dC = \frac{V_{O_2}^0}{n} \cdot \frac{C}{C_0} \cdot \frac{S \cdot n}{Q} dz \quad (6)$$

where S is the cross-sectional area of the coal sample (cm^2) and Q is the gas flow (mL/s).

When the temperature is constant, $V_{O_2}^0(T)$ and C_0 are constant, and the integral of eq 6 is as follows:

$$\int_{C_i}^{C_{i+1}} \frac{dC}{C} = - \frac{V_{O_2}^0(T) \cdot S}{Q \cdot C_0} \int_{C_i}^{C_{i+1}} dz \quad (7)$$

The following equation can be used to calculate the oxygen consumption rate:

$$V_{O_2}^0(T) = \frac{Q \cdot C_0}{S(Z_{i+1} - Z_i)} \ln \frac{C_i}{C_{i+1}} \quad (8)$$

According to eq 8 and the oxygen concentration in Figure 5, the variations of the coal oxygen consumption rate with the coal temperature under different air flow rates were calculated

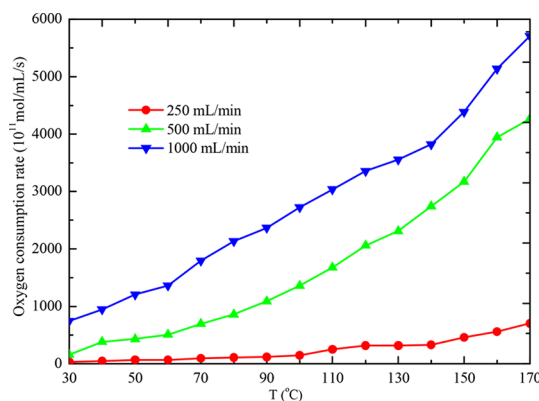


Figure 5. Oxygen consumption rate.

and are shown in Figure 5. The oxygen consumption rate increased as the temperature increased. In the air flow rate range 250–1000 mL/min, the higher the gas flow was, the higher the oxygen consumption rate became.

In the experimental tank, as the coal consumed oxygen, the oxygen concentration decreased along the airflow direction, while the CO concentration increased. The CO generation rate of coal at a certain point in the sample tank was directly proportional to the oxygen consumption rate, which is shown as follows:

$$V_{CO}^0(T) = \frac{C_{O_2}}{C_{O_2}^0} V_{CO}(T) \quad (9)$$

where $V_{CO}(T)$ is the rate of CO production (mol/ cm^3/s) and $V_{CO}^0(T)$ is the rate of CO production at the standard oxygen concentration (21%, mol/ cm^3/s).

The oxygen concentration at any point in the furnace can be deduced from Arrhenius' law, as follows:

$$C = C_i \cdot e^{-[V_{CO}^0(T) \cdot S / Q \cdot C_{O_2}^0] (z - z_i)} \quad (10)$$

$$dC_{CO} = \frac{V_{CO}(T)}{n} d\tau, \quad d\tau = \frac{dz}{u}, \quad u = \frac{Q}{S \cdot n} \quad (11)$$

The oxygen concentration at the high-temperature point is denoted as C_1 , and the distance from the inlet is Z_1 . Similarly, the oxygen concentration at the next point is C_2 , and the distance from the entrance is Z_2 . Substituting eq 10 into eq 11 and integrating yield the following:

$$\begin{aligned} C_{CO}^2 - C_{CO}^1 &= \int_{Z_1}^{Z_2} \frac{V_{CO}(T)}{u \cdot n} dz \\ &= \int_{Z_1}^{Z_2} \frac{S}{Q} \cdot \frac{C \cdot V_{CO}^0(T)}{C_{O_2}^0} dz \\ &= \frac{S \cdot V_{CO}^0(T)}{Q \cdot C_{O_2}^0} \int_{Z_1}^{Z_2} C_1 e^{-V_{CO}^0(T) \cdot S \cdot n \cdot (z - z_1) / Q \cdot C_{O_2}^0} dz \end{aligned} \quad (12)$$

According to eq 12, the CO generation rate at the standard oxygen concentration is obtained, as follows:

$$V_{CO}^0(T) = \frac{V_{CO}^0(T) \cdot (C_{CO}^2 - C_{CO}^1)}{C_1 \cdot [1 - e^{-[V_{CO}^0(T) \cdot S \cdot n / Q \cdot C_{O_2}^0] (z - z_1)}]} \quad (13)$$

Similarly, the rate of CO₂ generation is as follows:

$$V_{CO_2}^0(T) = \frac{V_{O_2}(T) \cdot (C_{CO_2}^2 - C_{CO_2}^1)}{C_1 \cdot [1 - e^{-[V_{O_2}(T) \cdot S \cdot n / Q \cdot C_0] (z_2 - z_1)}]} \quad (14)$$

Based on the CO concentration curve in Figure 3 and the oxygen consumption rate curve in Figure 5, the CO generation rate can be calculated using eq 13. The variation of the CO generation rate with temperature is shown in Figure 6. The CO

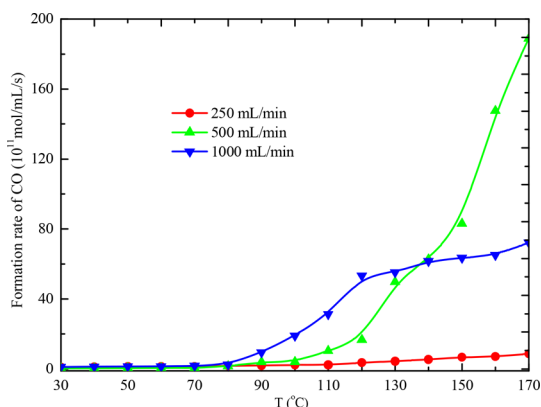


Figure 6. Generation rate of CO.

generation rate was low at low temperatures and did not change significantly with temperature. When the coal temperature was higher than 90 °C, the CO generation rate began to increase significantly with the increase in the coal temperature. However, there were significant differences in the CO generation rates in the high-temperature stage due to the different air volumes. In the temperature range 90–140 °C and the air flow rate range 250–1000 mL/min, the higher the air flow was, the higher the CO generation rate became. When the coal temperature was higher than 140 °C, the CO generation rate was the highest and increased rapidly at an air flow of 500 mL/min, followed by 1000 mL/min, and the lowest was 250 mL/min.

Similarly, based on the CO₂ concentration curves in Figure 4 and the oxygen consumption rate curves in Figure 5, the CO₂ generation rate could be calculated using eq 14. The variations of the CO₂ generation rate with temperature are shown in Figure 7. The results showed that when the coal temperature was lower than 130 °C, the CO₂ generation rate was basically the same under air flow rates of 250 and 500 mL/min. When the coal temperature was higher than 130 °C, there was a

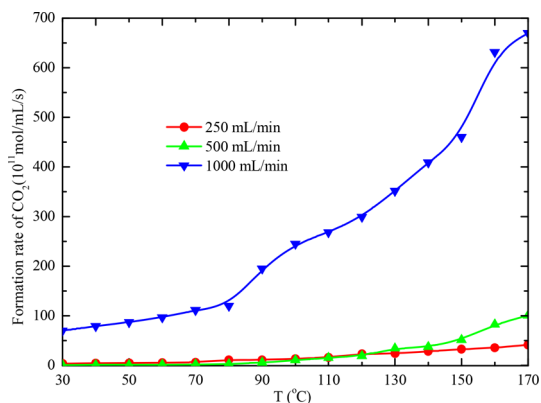


Figure 7. Generation rate of CO₂.

gradual difference between the two, and the CO₂ generation rate under the air flow rate 1000 mL/min was much higher than those at other air flow rates, indicating that the larger the air flow rate was, the more thorough the coal oxidation and spontaneous combustion were, and the higher the CO₂ generation rate became.

2.3. Analysis of Spontaneous Combustion Oxidative Exothermic Intensity. The calculation of the exothermic strength was based on the conservation of chemical bond energy, which included two kinds of exothermic strength calculations. One was the upper limit of the coal oxidative exothermic intensity ($q_{\max}(T)$). It was assumed that the oxygen consumption only generated CO and CO₂, so, the calculated heat release intensity was greater than the actual heat release intensity, which is called the upper limit of the coal oxidative exothermic intensity and can be calculated as follows:

$$q_{\max}(T) = \frac{V_{CO}^0(T)}{V_{CO}^0(T) + V_{CO_2}^0(T)} V_0(T) \cdot \Delta H^{CO} + \frac{V_{CO_2}^0(T)}{V_{CO}^0(T) + V_{CO_2}^0(T)} V_0(T) \cdot \Delta H^{CO_2} \quad (15)$$

For the other, it was assumed that all the forms of oxygen consumption other than CO and CO₂ occurred via chemisorption. The lower limit of the oxidative exothermic intensity ($q_{\min}(T)$) can be obtained as follows:

$$q_{\min}(T) = \Delta H^r \cdot [V_0(T) - V_{CO}^0(T) - V_{CO_2}^0(T)] + \Delta H^{CO} V_{CO}^0(T) + \Delta H^{CO_2} V_{CO_2}^0(T) \quad (16)$$

In the process of calculating the exothermic strength, according to refs 30 and 40, the chemisorption heat of coal was 58.8 kJ/mol, and the average thermal effect of oxidation generation of CO, CO₂, and H₂O was $\Delta H^{CO} = 319.5$ kJ/mol, $\Delta H^{CO_2} = 446.7$ kJ/mol, and $\Delta H^r = 348.6$ kJ/mol, respectively.

According to the conservation of chemical bond energy, there are two kinds of exothermic strengths: the upper and lower limits of the oxidative exothermic intensity. Two kinds of exothermic intensities have application value. As long as the same exothermic intensities are used in the comparative analysis, the analysis results are reliable and can be compared.⁴¹ Because the adiabatic spontaneous combustion period of coal is the shortest time to characterize its spontaneous combustion, and based on the guiding role of this index in engineering practice, the minimum value or lower limit of the calculated results can be selected within a reasonable calculation range to ensure safety. Therefore, we decided to use the upper limit of the coal oxidation exothermic intensity ($q_{\max}(T)$) as the basis for the comparative analysis of the oxidation exothermic intensity and the calculation of the spontaneous combustion period. Because the oxidation exothermic intensity was overestimated, the spontaneous combustion period calculated based on this was small, and thus, it would have higher reliability in guiding engineering practice.

According to eq 15, the oxygen consumption rate (Figure 5), the generation rate of CO (Figure 6), and the generation rate of CO₂ (Figure 7), the variations of the upper limits of the oxidation exothermic intensities of the coal samples were calculated and plotted versus temperature for three air flow conditions, as shown in Figure 8. The maximum upper limit of the oxidation exothermic intensity was at an air flow of 1000

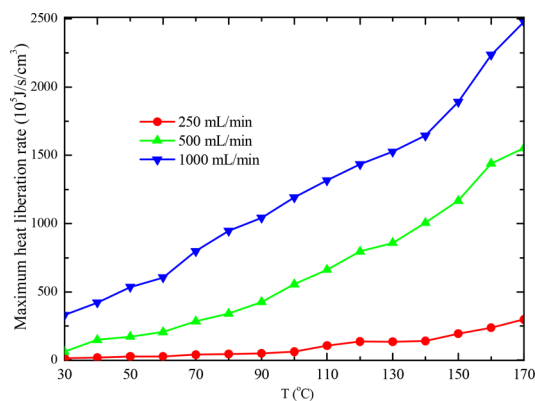


Figure 8. Upper limits of the oxidation exothermic intensity.

mL/min, the second was at 500 mL/min, and the lowest was at 250 mL/min. To facilitate subsequent research and calculation of the oxidation exothermic intensity at low temperatures, the three curves in Figure 8 were fitted, and the fitting results were as follows:

$$\begin{cases} q_{250\text{mL/min}} = 28.72 \exp(T/71.60) - 35.84 & R^2 = 0.99324 \\ q_{500\text{mL/min}} = 394.20 \exp(T/103.86) - 278.77 & R^2 = 0.99766 \\ q_{1000\text{mL/min}} = 497.36 \exp(T/99.03) - 278.77 & R^2 = 0.99476 \end{cases} \quad (17)$$

In the follow-up study, in the calculation of the adiabatic spontaneous combustion period, the corresponding exothermic intensity of the low-temperature oxidation was calculated using eq 17 and then used for relevant calculations and analysis.

2.4. Calculation Method and Results of the Coal Adiabatic Oxidation Natural Ignition Period. **2.4.1. Calculation Method of the Coal Adiabatic Oxidation Spontaneous Ignition Period.** During the experiment, the following phenomena actually exist. (1) As the content of nitrogen in air accounts for about 79%, and oxygen accounts for about 21%, and the nitrogen used in the experiment is 99.99% high-purity nitrogen, the difference in the thermal physical property parameters of air and nitrogen mainly lies in about 21% oxygen, so there is a certain difference in the thermal physical property parameters of air and nitrogen, but the difference is not significant. (2) The change of temperature is the root and key factor of the change of the coal pore structure influenced by the thermal effect. (3) There is no oxidation of coal samples in the nitrogen environment. Oxidation of coal in the air environment will consume a certain amount of oxygen and generate CO, CO₂, etc. However, since the coal sample is only about 1 kg, the absolute volume of oxygen consumed and gas produced is very small. Based on the above facts, and for the purpose of study of the spontaneous combustion period under adiabatic conditions, the following hypotheses were made based on the experimental research results of the temperature-programmed experiments in the air and N₂ environments.

(1) It was assumed that the heat capacities of nitrogen and air were the same and the gas flow velocities and temperatures of the air and nitrogen were the same when they flowed into the coal sample under the same pressure and flow path. (2) Under the influence of thermal effects, the pore structure of coal would change to some extent. It was assumed that at the same temperature, the thermal effects of air and nitrogen on

the pore structure of coal were the same. (3) Nitrogen was significantly different from the gases produced and released in an air environment. There were also significant differences in the gases produced and emitted under different gas flow rates in the same gas environment. However, the total amounts of gas generation and emission were relatively low and had little influence on the heat in the system. Therefore, the heat removed by different types and amounts of gases emitted under the same flow of air and nitrogen was ignored when they left the system.

In the process of temperature-programmed heating, the coal temperature under the air and nitrogen atmospheres changed with time, as shown in Figure 9. There were certain differences

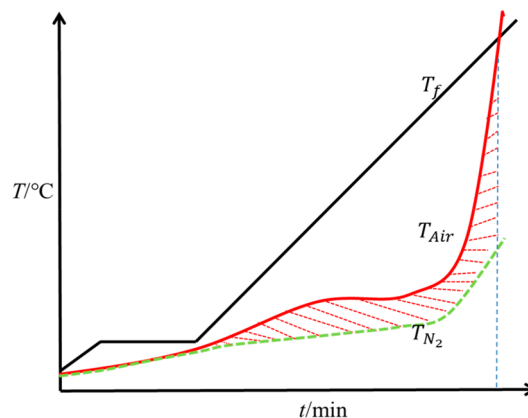


Figure 9. Temperature variations with time.

in the coal temperatures under the nitrogen and air atmospheres in the experimental process. The coal temperature differences mainly arose from the significant oxidation and spontaneous combustion under air conditions. The temperature differences between the air and nitrogen atmospheres were also nonadiabatic oxidation temperature differences, because under the temperature-programmed experimental conditions, the temperature of the experimental system was higher than the temperature of the coal. Thus, there was no heat loss from the coal sample to the outside. The term “adiabatic” means that neither heat dissipation nor absorption from the outside occur. Therefore, the temperature rise of the coal during spontaneous combustion in the air environment also was caused by two different heat sources: one was the chemical heating caused by the heat release from the oxidative spontaneous combustion, and the other was the physical heating of the furnace body and the heated airflow. In the comparative experiments with air and nitrogen, the temperature rise of the coal sample under the nitrogen atmosphere was caused by physical heating, and the temperature rise of the coal sample under the air atmosphere minus the physical heating component was caused by chemical heating due to oxidation. The coal temperature rise caused by chemical heating also included the oxidative reaction heat release promoted by physical heating; that is, oxidative heat release was promoted by external heat absorption. For pure adiabatic oxidation heat release, that is, the oxidation heat release of coal under conditions with no external heat absorption, the key to calculating the adiabatic natural combustion period is to determine the adiabatic oxidation heat release.

In Figure 10, the upper limit of the oxidation exothermic intensity is the curve shown in Figure 8. Under the

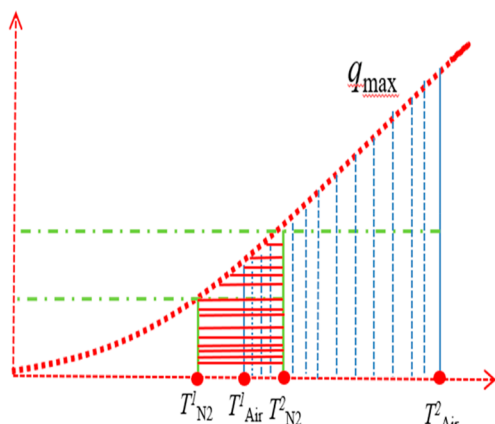


Figure 10. Exothermic strength variation with temperature.

temperature-programmed experimental conditions, all heat released in an air environment is the shaded area when the coal temperature increased from T_{Air}^1 to T_{Air}^2 . In the air environment, there was also oxidative heat release promoted by the physical heating of the furnace body and airflow, which was due to the exothermic oxidation reaction promoted by physical heating. The temperature corresponding to physical heating increased from $T_{N_2}^1$ to $T_{N_2}^2$. Therefore, the oxidative heat release promoted by external heat absorption caused by physical heating is the shaded area from $T_{N_2}^1$ to $T_{N_2}^2$ in Figure 10. The difference between the two shaded areas is the pure adiabatic oxidation heat release without any external heat absorption and release, expressed as follows:

$$\Delta q_{\max} = (q_{\max})_{T_{Air}^1}^{T_{Air}^2} - (q_{\max})_{T_{N_2}^1}^{T_{N_2}^2} \quad (18)$$

After the adiabatic oxidation heat release is known, the adiabatic heating rate Γ_o of the coal temperature promoted by adiabatic oxidation heat release can be calculated as follows:

$$\Gamma_o = \Delta q_{\max} / C \quad (19)$$

where C represents the heat capacity of the coal at different temperatures ($J \cdot g^{-1} \cdot K^{-1}$).

During the experiment, the gas composition and concentration were measured and analyzed every $10^\circ C$ from 30 to $170^\circ C$. The corresponding adiabatic heat release was calculated using the following equation:

$$\Delta q_{\max}(10^\circ C) = (q_{\max})_{T_{Air}^{i-1}}^{T_{Air}^i}(10^\circ C) - (q_{\max})_{T_{N_2}^{i-1}}^{T_{N_2}^i}(10^\circ C) \quad (20)$$

where T_{Air}^{i-1} is the lower boundary temperature corresponding to the $10^\circ C$ interval, T_{Air}^i is the upper boundary temperature corresponding to the $10^\circ C$ interval, $T_{N_2}^{i-1}$ and $T_{N_2}^i$ are the coal temperatures in the nitrogen environment, and T_{Air}^{i-1} and T_{Air}^i are the coal temperatures in the air environment.

The adiabatic oxidation heating rate corresponding to every interval of $10^\circ C$ in the air environment during the temperature-programmed experiment ($\Gamma_o^{(\Delta T_{Air}=10^\circ C)}$) can be calculated as follows:

$$\Gamma_o^{(\Delta T_{Air}=10^\circ C)} = \Delta q_{\max}(10^\circ C) / C_{T_{i-1}} \quad (21)$$

The adiabatic oxidation heating time corresponding to every interval of $10^\circ C$ (Δt_{oi-1}^i) in an air environment during the temperature-programmed experiment can be calculated as follows:

$$\Delta t_{oi-1}^i = \frac{10^\circ C}{\Gamma_o^{(\Delta T_{Air}=10^\circ C)}} \quad (22)$$

The adiabatic spontaneous combustion period (t) from 30 to $170^\circ C$ during the experiment can be calculated as follows:

$$t = \sum_1^i \Delta t_{oi-1}^i \quad (23)$$

2.4.2. Coal Adiabatic Spontaneous Combustion Period Results and Discussion. Based on the calculation method of the coal adiabatic spontaneous combustion period and eqs 18–22, the adiabatic oxidation heating times from 30 to $170^\circ C$ at intervals of $10^\circ C$ are listed in Tables 1, 2, and 3 under air inflows of 250 , 500 , and 1000 mL/min, respectively.

Based on Tables 1–3 and eq 23, the adiabatic spontaneous combustion periods of coal were 27.78 , 12.31 , and 10.44 d under air flows of 250 , 500 , and 1000 mL/min, respectively.

Table 1. Adiabatic Oxidation Calculation Results of Coal Samples under an Air Inflow of 250 mL/min

$T_{Air}/^\circ C$	$T_{N_2}/^\circ C$	$\Delta q_{\max}/10^5 \times J \cdot cm^{-3} \cdot s^{-1}$	$C_T/J \cdot g^{-1} \cdot K^{-1}$	$\Delta q_{\max}^i - \Delta q_{\max}^{i-1}/J \cdot g^{-1} \cdot h^{-1}$	$\Gamma_o/K \cdot h^{-1}$	$\Delta t_{oi-1}^i/h$
30	27.6	1.44	0.88			
40	35.7	2.93	0.91	0.07	0.08	132.5
50	44	4.64	0.94	0.08	0.08	118.8
60	52.6	6.52	0.97	0.09	0.09	111.9
70	61.3	8.74	1.00	0.10	0.10	97.8
80	69.5	11.98	1.03	0.15	0.15	68.9
90	73.8	20.44	1.06	0.39	0.37	27.1
100	76.9	32.01	1.09	0.53	0.49	20.4
110	80.7	44.83	1.12	0.59	0.53	18.9
120	85.7	58.43	1.15	0.63	0.55	18.3
130	91.4	73.56	1.18	0.70	0.59	16.9
140	92	99.15	1.21	1.18	0.98	10.2
150	93.2	127.82	1.24	1.32	1.07	9.4
160	94.4	161.02	1.27	1.53	1.21	8.3
170	95.1	200.19	1.30	1.81	1.39	7.2

Table 2. Adiabatic Oxidation Calculation Results of Coal Samples under an Air Inflow of 500 mL/min

$T_{Air}/^{\circ}\text{C}$	$T_{N_2}/^{\circ}\text{C}$	$\Delta q_{max}/10^5 \times \text{J}\cdot\text{cm}^{-3}\cdot\text{s}^{-1}$	$C_T/\text{J}\cdot\text{g}^{-1}\cdot\text{K}^{-1}$	$\Delta q_{max}^i - \Delta q_{max}^{i-1}/\text{J}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	$\Gamma_o/\text{K}\cdot\text{h}^{-1}$	$\Delta t_{oi-1}^i/\text{h}$
30	29.6	2.02	0.88			
40	39.3	3.89	0.91	0.09	0.09	105.5
50	48.9	6.72	0.94	0.13	0.14	72.0
60	57.5	16.71	0.97	0.46	0.48	21.0
70	66.2	27.79	1.00	0.51	0.51	19.6
80	75.1	39.25	1.03	0.53	0.51	19.5
90	84.1	51.78	1.06	0.58	0.55	18.3
100	92.4	72.86	1.09	0.97	0.89	11.2
110	100.7	97.37	1.12	1.13	1.01	9.9
120	107.6	140.87	1.15	2.01	1.75	5.7
130	114.3	193.37	1.18	2.42	2.05	4.9
140	116.7	304.97	1.21	5.15	4.26	2.3
150	118.9	432.40	1.24	5.88	4.74	2.1
160	119.7	591.71	1.27	7.35	5.79	1.7
170	120.6	766.81	1.30	8.08	6.22	1.6

Table 3. Adiabatic Oxidation Calculation Results of Coal Samples under an Air Inflow of 1000 mL/min

$T_{Air}/^{\circ}\text{C}$	$T_{N_2}/^{\circ}\text{C}$	$\Delta q_{max}/10^5 \times \text{J}\cdot\text{cm}^{-3}\cdot\text{s}^{-1}$	$C_T/\text{J}\cdot\text{g}^{-1}\cdot\text{K}^{-1}$	$\Delta q_{max}^i - \Delta q_{max}^{i-1}/\text{J}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	$\Gamma_o/\text{K}\cdot\text{h}^{-1}$	$\Delta t_{oi-1}^i/\text{h}$
30	29.3	4.74	0.88			
40	39.1	6.74	0.91	0.09	0.10	98.8
50	48.8	9.93	0.94	0.15	0.16	63.9
60	57.8	20.03	0.97	0.47	0.48	20.8
70	66.5	35.02	1.00	0.69	0.69	14.5
80	75.2	52.78	1.03	0.82	0.80	12.6
90	83.2	81.90	1.06	1.34	1.27	7.9
100	84.3	200.17	1.09	5.46	5.01	2.0
110	86.1	323.86	1.12	5.71	5.10	2.0
120	87.7	465.02	1.15	6.51	5.67	1.8
130	88.9	627.86	1.18	7.52	6.37	1.6
140	90.6	803.12	1.21	8.09	6.69	1.5
150	92.3	998.89	1.24	9.04	7.29	1.4
160	94	1217.37	1.27	10.08	7.94	1.3
170	95.3	1466.29	1.30	11.49	8.84	1.1

The adiabatic spontaneous combustion periods for 250 and 1000 mL/min air flow rates were 2.26 and 1.18 times that of the 500 mL/min air flow rate, respectively. In the process of the air flow rate increasing from 250 to 1000 mL/min, the adiabatic spontaneous combustion period was shortened, and as the air volume increased, the difference between the adiabatic spontaneous combustion periods decreased.

Yang et al.⁴² found that the coal spontaneous combustion period decreased first and then increased with the increase in the air leakage velocity, which verified that the research results of the present experiment were reasonable. According to Yang's research results and the variation behaviors of the adiabatic spontaneous combustion period under different air flow rates obtained in this paper, there is a quadratic function relationship between the adiabatic spontaneous combustion period and the air flow rate, expressed as follows:

$$t = 0.2791q^2 - 7.2012q + 52.94 \quad (R^2 = 1) \quad (24)$$

where t is the adiabatic spontaneous combustion period (d) and q is the air flow (mL/s).

According to eq 24, when the air flow rate q was 12.9 mL/s (774 mL/min), the adiabatic spontaneous combustion period of the lignite used in this experiment was 6.5 d. This means that if adiabatic oxidation experiments were used to obtain the adiabatic spontaneous combustion period of this coal, the

shortest time to carry out the experiment would be 6.5 d, which would be too long to obtain rapid results. This is one of the advantages of the proposed experimental method.

The adiabatic spontaneous combustion period of coal is the result of multiple external factors. In the research process described in this paper, we found that during the coal spontaneous combustion experiments, the furnace body and the heated air also had significant physical heating effects on the coal, and the change of the coal pore structure, water migration, and decomposition of the functional groups were highly related to the temperature rise of the coal. In the experimental process, if noncoal samples were used as control samples or noncoal samples were compared with coal, the effect of temperature on the above factors would be ignored. This would result in significant differences between the spontaneous combustion process of coal studied in the experiment and the actual process of coal spontaneous combustion, and the characteristics of coal spontaneous combustion would not be accurately reflected. In this paper, the same coal was used for the experimental samples, and pure nitrogen and air were used to carry out the control experiments. The oxidizing medium of the coal was varied, but the characteristics of the coal itself were not changed, and the spontaneous combustion process of the coal was accurately reflected.

Various issues must be clarified by integrating the experiment and calculation processes to improve the method proposed in this paper. First, the test of each gas component and concentration in the temperature-programmed experiment must be accurate. Second, the heat of chemical adsorption of coal and the heats of formation of CO, CO₂, and H₂O must be true and accurate.

3. CONCLUSION

In summary, this study established a rapid contrastive experimental method to quickly obtain the accurate adiabatic spontaneous combustion period of coal. The study illustrated that in the temperature-programmed coal spontaneous combustion experiments, the increase in the coal temperature was due to physical and chemical heating, and the chemical heating included oxidative exothermic heating promoted by physical heating and adiabatic oxidation heating. The adiabatic spontaneous combustion period of coal was calculated from the adiabatic oxidation heat release. The adiabatic spontaneous combustion period of coal under different air flow rates was in good agreement with existing research results, and the differences between existing research results were explained. This rapid contrastive experimental method can greatly shorten the experimental time and completely and accurately reflect the influence of the coal pore structure, water migration, and functional group decomposition on spontaneous combustion. This method provides a new approach to quickly and accurately determine the adiabatic spontaneous combustion period of coal based on the conservation of chemical bond energy. The main conclusions are as follows:

1. In the process of the temperature-programmed experiment of spontaneous combustion of coal in an air environment, there are two heat sources for the temperature rise of coal. The first is the physical heat source composed of the experimental furnace body and the air flow after heating. The second is the total heat released by the oxidation reaction of coal and oxygen in the temperature-programmed process, which is the chemical heat source or oxidation reaction heat. In the experimental process, the total heat released by the oxidation reaction of coal samples, namely the chemical heat source, also includes the reaction heat released by accelerated oxidation caused by physical heating and the adiabatic oxidation heat release. Adiabatic oxidation heat release is the key basis for calculating the adiabatic combustion period of coal. If the adiabatic oxidation heat release can be determined, the adiabatic combustion period can be determined.
2. The calculation method of the adiabatic oxidation heat release and adiabatic spontaneous combustion period of coal based on a self-designed rapid experimental system is presented. The first step is to establish a functional relationship between the heat releasing intensity and temperature in an air environment. The second step is to calculate the oxidative heat release intensity accelerated by the physical heat source, according to the temperature change of the coal sample under a nitrogen environment, by using the relation function of heat release intensity and temperature. In the third step, the adiabatic oxidative heat release intensity is obtained by subtracting the oxidative heat release intensity accelerated by physical heating calculated in the second step

from the oxidative exothermic intensity in the air at the same time. Finally, the adiabatic spontaneous combustion period of coal is calculated by summing the adiabatic oxidation times of different temperature stages.

3. This rapid contrastive experimental method can greatly shorten the experimental time and can completely and accurately reflect the influence of the coal pore structure, water migration, and functional group decomposition on spontaneous combustion. At the same time, it can ensure that the experimental and solving process of the adiabatic spontaneous combustion period of coal are real adiabatic processes.

4. MATERIALS AND METHODS

4.1. Materials. The samples of lignite in this experiment were obtained from the Fengshuigou Coal Mine of the

Table 4. Results of Proximate Analysis of Coal Samples

M(ad)/%	A(ad)/%	V(ad)/%	FC(ad)/%
12.26	9.82	33.13	44.81

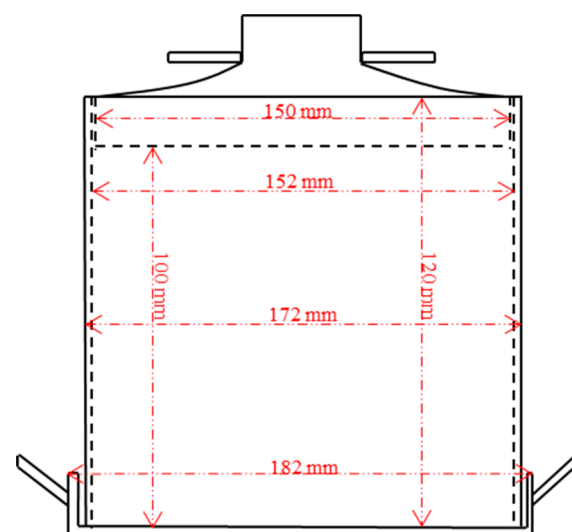


Figure 11. Coal sample crushing tank.

Table 5. Particle Size Composition of the Sample

10–20 mm	5–10 mm	2.5–5 mm	1.0–2.5 mm	0.5–1.0 mm	0.25–0.5 mm	<0.25 mm
30.10	28.77	13.63	15.27	5.93	3.60	2.77

Pingzhuang Coal Company in Inner Mongolia, China. The proximate analysis results are shown in Table 4, where $M(ad)$ is the moisture content of the coal sample, $A(ad)$ is the ash content of the coal sample, $V(ad)$ is the volatile content of the coal sample, and $FC(ad)$ is the fixed carbon content of the coal sample. The uniaxial compressive strength of the coal was 4.87–5.66 MPa, and the tensile strength was 0.93 MPa.

4.2. Sample Preparation and Temperature-Programmed Experimental Method. **4.2.1. Sample Preparation.** Fresh coal samples were collected from the coal mine working face of the Fengshuigou Coal Mine and sealed with plastic film. Under the action of mechanical grinding, coal oxidizes and forms a large number of oxygen-containing functional groups. The effect of mechanical grinding on the

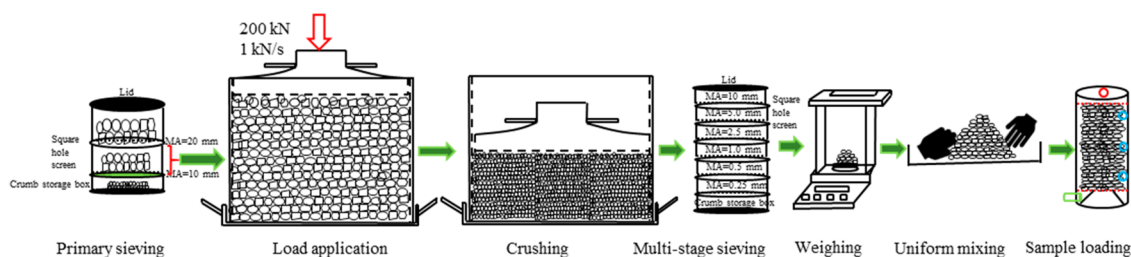
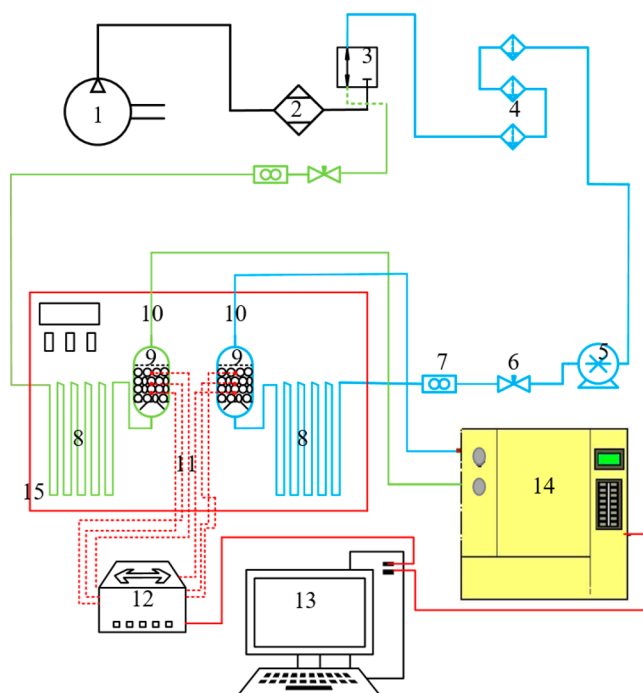


Figure 12. Coal sample preparation process.



1. Air compressor, 2. refrigerated air dryer, 3. three-way ball valve, 4. three-stage high-efficiency particulate air filter (HEPA), 5. nitrogen generator, 6. ball valve, 7. gas flow meter, 8. fine copper tube, 9. pure copper coal sample tank and coal sample, 10. tail pipe, 11. K-type temperature sensor and data transmission line, 12. multichannel temperature inspection instrument, 13. computer monitor, 14. gas chromatograph, and 15. temperature-programmed furnace

Figure 13. Temperature-programmed experimental system.

spontaneous combustion of coal is caused not only by the change of the specific surface area but also by the mechanochemical action during grinding.^{37–39} Thus, the coal was crushed instead of ground for these experiments.

To ensure that the coal samples used in the experiment were as consistent with the coal crushing process in the goaf as possible, the sealed coal samples were broken manually with a hammer in the laboratory. Bulk coal with a particle size of 10–20 mm was selected, and it was placed in a crushing tank (as shown in Figure 11). According to the compressive strength of the coal sample and the maximum stress concentration in a goaf, the coal sample in the coal sample tank was loaded at a speed of 1 kN/s and a maximum pressure of 200 kN. It was unloaded after reaching the maximum load of 5 s. The crushed coal samples were removed, sieved, and weighed, and the mass ratios of coal samples with different particle sizes were evaluated. The particle sizes and corresponding mass percentages of the crushed coal samples are shown in Table 5. The samples were evenly mixed and placed in a copper coal sample tank. The coal sample preparation process is shown in

Figure 12. The mass of each coal sample in the copper coal tank m was 1200 g, the volume V was 1583 mL, the height H was 20 cm, and the porosity n was 0.56.

4.2.2. Temperature-Programmed Experimental Method. The temperature-programmed experimental system shown in Figure 13 was established. This system could carry out temperature-programmed tests of the coal in nitrogen and air simultaneously. Two coal sample tanks were in the temperature-programmed box. One tank was connected to a nitrogen pipeline, and the other was connected to an air pipeline. Exhaust emission pipelines were connected to both tanks. Temperature sensors were connected, and the gas path and coal sample tank were finally sealed. The temperature-monitoring systems were enabled. First of all, connect the two coal sample tanks with the nitrogen pipeline, turn on the nitrogen, and turn on the heating program, starting with room temperature and using a heating rate of 1 °C/min; when the coal temperature reaches 30 °C, stop heating. Start the thermostatic procedure, at the constant temperature 60 min; after the constant temperature, stop the nitrogen supply. The

nitrogen pipe of the coal sample can disconnect from the coal sample tank of the air atmosphere, connect the nitrogen and air pipelines, respectively, to the two coal samples, and ensure the same gas flow of the two coal sample tanks. Start the heating program again at a heating rate of 1 °C/min, and stop the heating when the temperature of the coal sample reaches 170 °C in the air environment. Gas samples were collected and analyzed by gas chromatography as the temperature was varied in an air atmosphere from 30 °C to the maximum temperatures of the coal samples at intervals of 10 °C to determine the compositions and concentrations of the gas samples. The experiment was stopped when the temperatures of the coal samples in the air environment reached 170 °C.

AUTHOR INFORMATION

Corresponding Author

Ziwen Dong – School of Safety Engineering, Ningbo University of Technology, Ningbo, Zhejiang 315211, China;
orcid.org/0000-0002-4926-9380; Email: 1316859454@qq.com

Authors

Lifeng Sun – Emergency Research Institute, China Coal Research Institute CCRI, Beijing 100013, China
Tingguai Jia – Institute of Mining and Coal, Inner Mongolia University of Science & Technology, Baotou, Inner Mongolia 014010, China
Shengli Guo – School of Energy and Safety, Anhui University of Science & Technology, Huainan 232001, China
Weile Geng – School of Resources and Safety Engineering, Chongqing University, Chongqing 400030, China
Wenhui Yu – School of Safety Engineering, Ningbo University of Technology, Ningbo, Zhejiang 315211, China
Zikun Pi – School of Safety and Environment Engineering, Hunan Institute of Technology, Hengyang, Hunan 421002, China
Guangli Li – School of Safety and Environment Engineering, Hunan Institute of Technology, Hengyang, Hunan 421002, China
Chen Luo – School of Safety and Environment Engineering, Hunan Institute of Technology, Hengyang, Hunan 421002, China

Complete contact information is available at:

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Notes

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