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Evaluation and Optimization of Multi-Parameter Prediction Index for Coal Spontaneous Combustion Combined with Temperature Programmed Experiment

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Abstract: Coal spontaneous combustion (CSC) is a serious threat to the safe mining of coal resources, and the selection of suitable gas indicators to predict the CSC state is crucial for the prevention and control of coal mine fires. In this paper, the temperature-programmed experiment of CSC was first carried out to analyze the gas components and compositions in the oxidative pyrolysis process of three coal samples (lignite, long-flame coal, and lean coal) with different coalification degrees. Subsequently, the spontaneous combustion tendency of these three coal samples was evaluated. Finally, through the variation of gas concentration, gas concentration ratio, and fire coefficient with coal temperature, the indicators suitable for predicting the spontaneous combustion of coal were preferred, and a multi-parameter indicator system was established to make a comprehensive judgment on the spontaneous combustion status of coal. The results show that coal rank is negatively correlated with oxygen consumption rate. The higher the coalification degree of coal, the slower the oxidation reaction and the later the characteristic temperature point appears. The lignite selected in this experiment is a type of coal that is more prone to spontaneous combustion than long-flame coal and poor coal, and the CO concentration, C_2H_6/CH_4, and second fire coefficient R_2 can be used as the main indicators for predicting CSC, while the other gases, olefin-alkane ratio and fire coefficient can be used as auxiliary indicators. To some extent, the research content can effectively and accurately determine the stage and degree of coal spontaneous combustion, which has a certain guiding role in predicting CSC.

Keywords: coal spontaneous combustion; temperature programmed system; indicator gases; forecast indicators; fire coefficient

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1. Introduction

During coal mining, coal spontaneous combustion (CSC) disasters is often accompanied, which not only causes waste of coal resources, but also seriously threatens the safety of workers and coal production [1–5]. Coal spontaneous combustion (CSC) is a multivariate self-accelerating exothermic process, which is mainly the result of the compounding of coal and oxygen molecules [6,7]. Different gases are produced at different stages of the coal auto-ignition process, and there are certain laws between the gas types and concentrations and the coal temperature. These laws can be utilized to predict the state of different stages of coal auto-ignition [8–10]. Therefore, an in-depth understanding of the relationship between gas products and temperature in the process of coal self-heating is of great significance for fire prevention as well as for ensuring the safe development of the coal industry.

Scientists have carried out a special study on the gas produced during theCSC process to estimate the degree of coal oxygen reaction and the stage of coal spontaneous combustion in goaf. At the low-temperature stage, the generation of CO and CO_2 is generally considered to be caused by the direct decomposition of stable oxygen-containing complexes containing intrinsic oxygen-containing functional groups and a small number of combustion reactions. At the high-temperature stage, coal has a violent oxidation reaction, and more olefin and alkane gases are generally produced by pyrolysis [11]. In addition, accurate measurement of the reduction of oxygen concentration can also be applied to determine the spontaneous combustion state of coal [12]. Some scholars believe that some single gases, such as CO, CO_2, C_2H_4, and C_2H_6, can be used as indicator gases for predicting spontaneous combustion [4,13–15]. Subsequently, the CO/CO_2 ratio, C_2H_4/C_2H_6 ratio, $\Delta CO/\Delta CO_2$ ratio, and C_2H_6/C_2H_2 ratio are also considered to be able to predict spontaneous combustion of coal [16–18]. Based on these indicator gases, other scholars have proposed the Graham fire coefficient [Wangcaiping2021], Willet's ratio [19], C/H ratio [20], and Graham's ratio [21] to predict the self-heating state of coal. Zhao et al. pointed out that the concentration of CO and CO_2 fluctuated with the change of ventilation, and the reliability of the gas as an indicator of spontaneous combustion of coal was poor [22]. To sum up, there are many indicators to distinguish coal's spontaneous combustion status at present, but the prediction indicators are different in different environments. It is easy to use wrong indicators to predict coal spontaneous combustion, resulting in misjudgment. Coal spontaneous combustion is a complex physical and chemical reaction process, and gases will affect each other. A single index gas cannot effectively predict coal spontaneous combustion [23,24]. Therefore, it is necessary to establish a multi-parameter index system to fully and accurately understand the state and development of CSC.

This paper analyzes the types and concentrations of gases produced during the oxidation of lignite, long-flame coal, and lean coal based on temperature-programmed experiments and shows the variation of gas concentration ratios with temperature. Then, the spontaneous combustion tendency of these three coal samples was characterized. Combined with the variation law of three parameters, namely, index gas concentration, ratio of gas concentration and fire coefficient, with coal temperature, the indexes suitable for predicting spontaneous combustion of coal are optimized. A multi-parameter index system was established to evaluate the spontaneous combustion status of coal more accurately and reasonably.

2. Experiment

2.1. Materials

Three coal samples of different coalification degrees were selected from coal mines in China, with coal ranks ranging from low to high being, lignite, long-flame coal, and lean coal.

Fresh coal samples were collected from the coal mining face and sealed and packed for transportation back to the laboratory. After that, the oxidized layer on the surface of the coal samples was peeled off, and then each type of coal was crushed by a jaw crusher and sieved to find enough coal samples to be used in the experiments. The coal particles were then dried in a drying oven. Proximate analysis and ultimate analysis experiments of each coal sample were carried out to understand the coal quality characteristics of the three coal samples. The results are shown in Table 1.

Table 1. Proximate analysis and ultimate analysis of coal.

No.	Coal Rank	Proximate Analysis				Ultimate Analysis				
		M_{ad} (%)	A_d (%)	V_{daf} (%)	FC_{ad}	C	O	N	H	S
1#	Lignite	26.35	10.86	40.21	67.19	65.12	23.28	1.67	4.06	1.34
2#	Long-flame coal	10.23	9.14	31.73	75.93	72.48	17.43	1.52	5.12	0.72
3#	Lean coal	8.19	7.05	18.62	78.69	75.95	10.06	1.89	5.61	1.03

Note: M_{ad} is the moisture (air drying basis); A_d is the ash (dry basis); and V_{daf} is the volatile (dry ash-free); FC_{ad} is fixed carbon.

Among them, the moisture, volatile, and ash content of lignite are higher than the other two coal samples. Lignite has the highest oxygen content and the lowest carbon and hydrogen content. The types and contents of minerals in coal samples with different coalification degrees show obvious differences, and the differences in minerals in coal also have a certain effect on the low-temperature oxidation of coal. The particle size of coal sample required for the temperature-programmed test is 0.18~0.38 mm. Before the experiment, coal particles were weighed with a balance and placed in a drying environment of 40 °C for 48 h. After that, they were taken out and put into sealed bags for later use. The mass of coal samples in each group of experiments was 50 g. Prepare nitrogen cylinders of 99.99% purity and standard oxygen cylinders of 20.96% standard oxygen concentration for the experiment.

2.2. Methods

In this work, the oxidation characteristics of coal auto-ignition were tested using a temperature-programmed test. The temperature-programmed experiment system consists of three parts: an air supply system, a programmed heating system, and a gas analysis system. The air supply system contains compressed air bottles, pressure-reducing valves, flowmeters, pressure gauges, etc., which are connected to each other in turn by latex tubes. The temperature-programmed system consists of a thermostat and a temperature-programmed control device, which is equipped with a preheating tube and a coal sample tank. The temperature accuracy is controlled at about 0.1 °C. The gas analysis system consists of a gas chromatograph and a data analysis device. The specific temperature-programmed experimental system is shown in Figure 1.

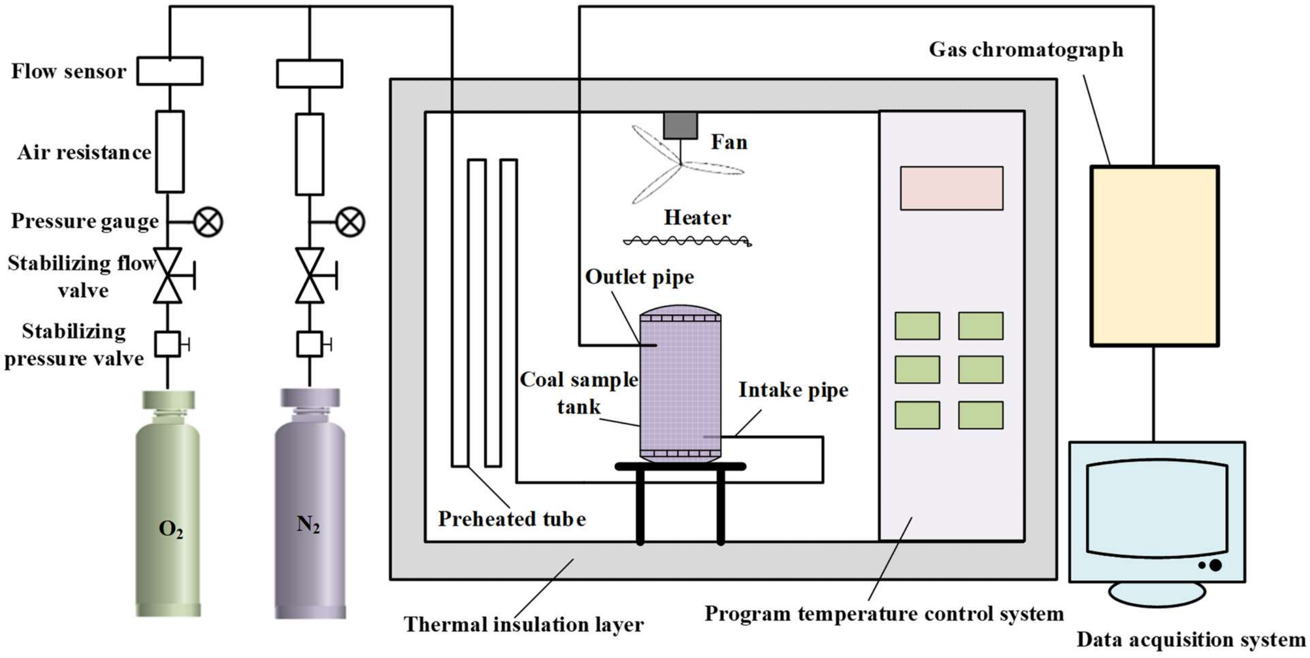

Figure 1. Temperature-programmed experimental system.

In a temperature-programmed oxidation experiment environment, the oxidation processing of coal itself will release heat, and the heat accumulation will lead to an increase in coal temperature [25,26]. Then, the experimental data are recorded by instrument, and the variation laws of coal temperature, the output of marker gas, and oxygen consumption with temperature are studied. Finally, the stage of spontaneous combustion to which the coal belongs is inferred, and early prediction of coal natural ignition is carried out. The main operation process of temperature programmed experiment can be summarized as follows:

- After checking and ensuring good gas tightness and proper functioning of all instruments, coal samples are placed into the coal sample tank, and a layer of asbestos with a thickness of 2~3 mm above the coal samples is evenly applied to prevent blocking of the gas path.
- The gas flow required for the experiment was adjusted; that is, dry air (oxygen concentration: 20.96%) was injected into the coal sample tank at a steady flow rate of 100 mL/min.
- The temperature is set through the temperature control system. The initial temperature is set to 30 °C, and the heating rate is 1 °C/min until it is heated to 200 °C. Here, ensure that the heating rate of each experiment is the same so as to improve the comparability of experimental results. The temperature sensor is used to monitor tank temperature and coal temperature, respectively, and the two are compared to obtain the crossing point temperature.
- The gas composition and concentration at different temperatures are monitored by gas chromatograph. We monitor the outlet gas concentration and the gas concentration produced by pyrolysis every 10 °C increment in temperature. Finally, the experimental results are recorded and saved in the computer terminal of the data collection system.

3. Results and Analysis

3.1. Outlet Oxygen Concentration and Oxygen Consumption Rate

Three coal samples with different degrees of deterioration were tested for oxidation at elevated temperatures, and their outlet oxygen volume fractions are shown in Figure 2a. Then, the oxygen consumption rate of each coal sample was calculated according to the calculation formula of oxygen consumption rate, as shown in Figure 2b.

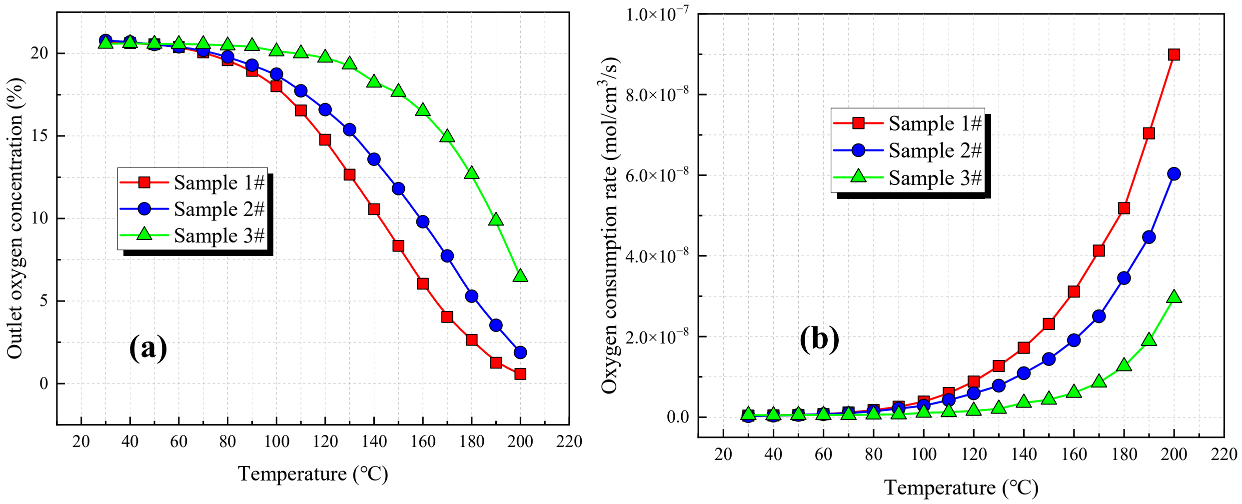

Figure 2. Variation of outlet oxygen concentration and oxygen consumption rate with temperature (**a**) outlet oxygen concentration (**b**) oxygen consumption rate.

It can be seen from Figure 2a that the oxygen concentration at the outlet of coal samples with different coalification degrees decreases with the increase in temperature. This indicates that the higher the temperature is, the more oxygen is involved in the reaction and the more violent the reaction is. When the temperature is fixed, the oxygen concentration at the outlet of the coal sample tank decreases with the increase in the coal rank. This indicates that the lower the rank of coal, the more intense the oxidation reaction of coal. In addition, Figure 2b more clearly shows that the oxygen consumption rate is positively correlated with temperature and negatively correlated with coal rank [22,27,28]. From the analysis of moisture, the moisture of each coal sample decreases with the increase in coalification degree. The water adsorbed in the pores inside the coal particles can evaporate only after the temperature reaches more than 100 °C and after a period of time. When the water evaporates, more pore structures in the coal are exposed to the air, accelerating the consumption of oxygen. Therefore, the higher the moisture content of the coal sample, the faster the oxygen consumption rate to some extent.

3.2. Gas Product Concentration

3.2.1. CO and CO_2

In the coal mining face, CO gas is released in large amounts and has good sensitivity, so CO is often selected as the index gas to divide different stages of the low-temperature oxidation process of coal [3,29]. Here, the variation of CO and CO_2 concentrations generated during the oxidation process is statistically shown in Figure 3. With the increase in temperature, CO and CO_2 concentrations of coal samples with different coalification degrees show an increasing trend with the increase in temperature. At the initial temperature of 30 °C, CO was detected in all kinds of coal, indicating that the coal had been slowly oxidized with O_2 at this time, and the CO gas was present throughout the whole process of temperature increase and oxidation. There is a sudden change in the process of CO production with the increase in temperature. This abrupt temperature point is the critical temperature of each coal sample, which is 60 °C, 70 °C, and 80 °C from lignite, long-flame coal, and lean coal. It can be observed that the higher the degree of coal deterioration, the greater the critical temperature point. The coal samples with high rank will make the critical temperature point that appears in the process of heating and oxidizing lag behind.

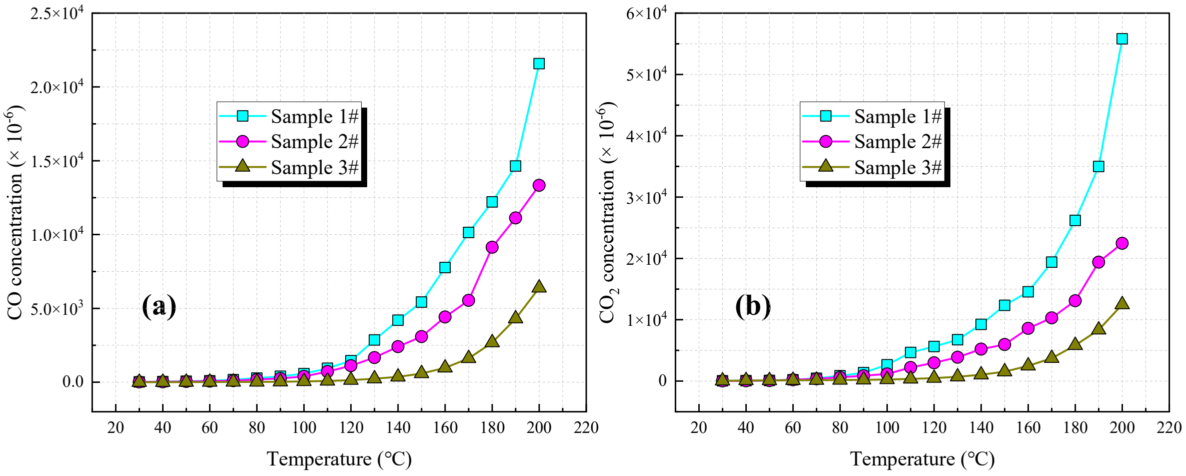

Figure 3. Variation of CO and CO_2 concentration with temperature (**a**) CO (**b**) CO_2.

3.2.2. CH_4 and C_2H_6

The variation trend of CH_4 and C_2H_6 gas concentration generated by temperature-programmed oxidation of each coal sample with temperature is shown in Figure 4. Overall, the CH_4 and C_2H_6 concentrations of each coal sample increase with the increase in temperature. Only the temperature points at which the two gases are generated are different. CH_4 is always present at all stages of temperature rise, which is due to the fact that the coal itself contains a small amount of CH_4 gas. The temperature points at which C_2H_6 gas is generated from lignite, long-flame coal, and lean coal are 90 °C, 100 °C, and 120 °C, respectively. The appearance of these characteristic temperature points can be considered that the spontaneous combustion and oxidation process of coal has entered an accelerated stage. At the same time, it also confirms the law that the higher the deterioration degree of coal, the more lagging the characteristic temperature. In addition, it can be seen from Figure 4a that the CH_4 concentration of Sample 2# of long-flame coal is higher than that of the other two coal samples because the CH_4 content adsorbed in the coal sample is higher. This presents that the CH_4 concentration under different coalification degrees and different environments is also quite different, and it is not appropriate to use a single CH_4 gas to predict the CSC process [9].

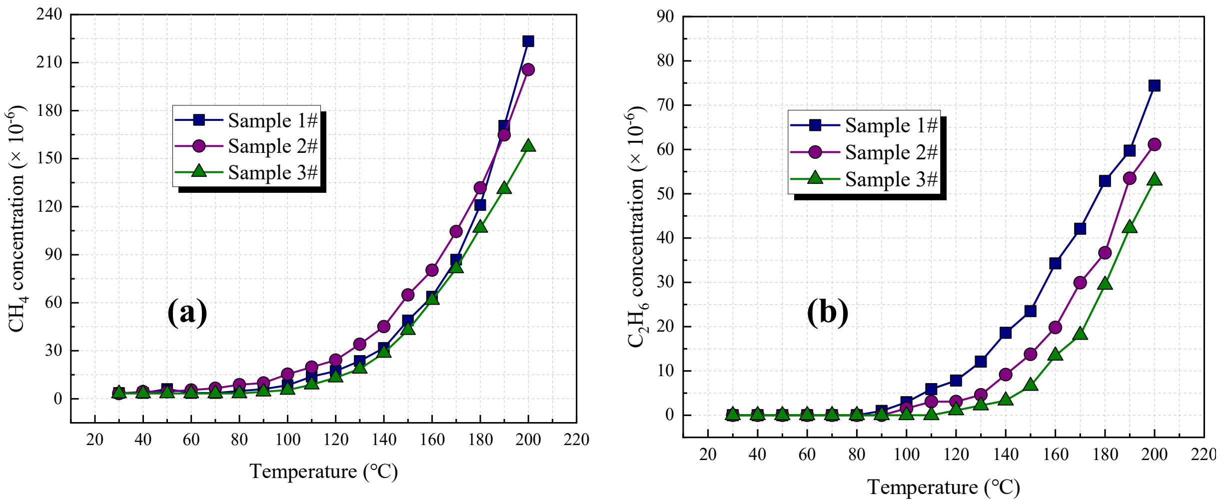

Figure 4. Variation of CH_4 and C_2H_6 concentrations with temperature (**a**) CH_4 (**b**) C_2H_6.

3.2.3. C_2H_4

Figure 5 illustrates the variation of the olefin gas C2H4 produced during spontaneous combustion of coal with temperature. It can be observed that C_2H_4 cannot be monitored at normal temperature, and the presence of C_2H_4 can be monitored only when a certain temperature value is reached. The temperature of C_2H_4 in coal samples with different coalification degrees is different. The temperature points of C_2H_4 gas generated by lignite, long-flame coal, and lean coal are 110 °C, 120 °C, and 150 °C, respectively. These temperatures can be referred to as dry cracking temperatures and symbolize the entry of the coal samples into the accelerated oxidation stage. After these temperature points, the C_2H_4 gas of each coal sample shows an exponential growth trend with the increase in temperature. For lignite and long-flame coals, a turning point in C_2H_4 concentration with increasing temperature occurs when the temperature is at 160 °C when the rate of C_2H_4 production becomes greater, and the oxidation reaction is more intense. For lean coal, the temperature at this transition point is about 180 °C. If this phenomenon is found, timely measures need to be taken to prevent the further expansion of spontaneous combustion.

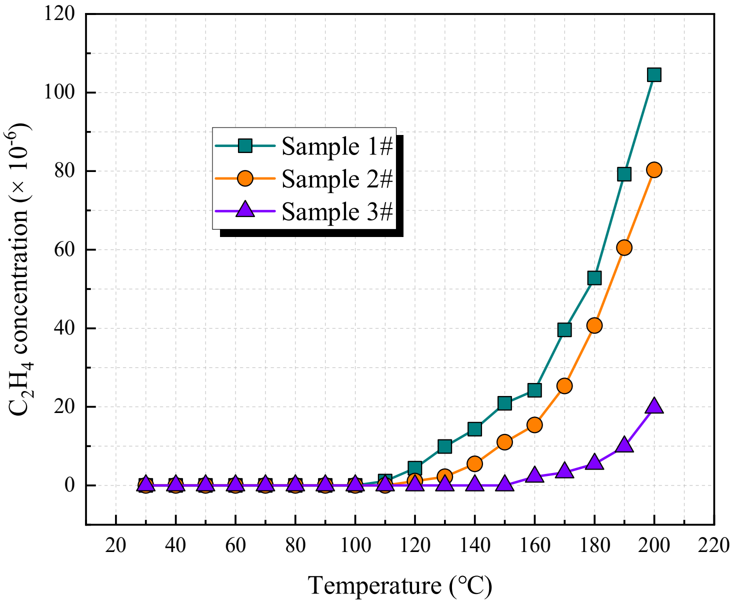

Figure 5. Variation of C_2H_4 concentration with temperature.

3.3. Change of Product Gas Concentration Ratio

The above only analyzes the change law of single index gas, and it is unreliable to predict the degree of coal spontaneous combustion only according to this [12,30]. Therefore, it is necessary to investigate the variation law of some composite index gases. The composite index gas here mainly refers to the ratio between the concentrations of main gases released during the natural oxidation and heating process of coal, including CO_2/CO (α), C_2H_6/CH_4 (β), C_2H_4/C_2H_6 (γ), and C_2H_4/CH_4 (δ).

3.3.1. CO_2/CO

Figure 6 shows the variation law of CO_2/CO (α) with temperature. The CO_2/CO of coal samples with different coalification degrees are different. For long-flame coal, the CO_2/CO of the whole temperature range is less than 5. Before about 50 °C, α decreases with the increase in temperature, indicating that the rate of CO production in the low-temperature oxidation stage is greater than that of CO_2. At this stage, the coal sample is in the slow oxidation stage, and the generated CO and CO_2 gases are less. Between 50 °C and 110 °C, α increases with the increase in temperature, indicating that the production rate of CO_2 is greater than that of CO. At this stage, the oxidation reaction rate of coal is increasing. After 110 °C, α decreases with the increase in temperature, and the production rate of CO is greater than that of CO_2. Coal spontaneous combustion enters an accelerated stage, and the oxidation reaction is intense. Lignite also shows a similar law with long-flame coal, but the characteristic temperature points of lignite should be compared with long-flame coal in advance. Additionally, for lean coal, α increases first and then decreases with the increase in temperature. However, this increasing trend is only in the short temperature stage of 30–40 °C. This is due to the higher rank of lean coal and better pore development, in which some CO_2 gas is adsorbed. At the beginning of heating up, CO_2 is desorbed out in a short time, resulting in the production rate of CO_2 being greater than that of CO. After 40 °C, the production rate of CO_2 is lower than that of CO.

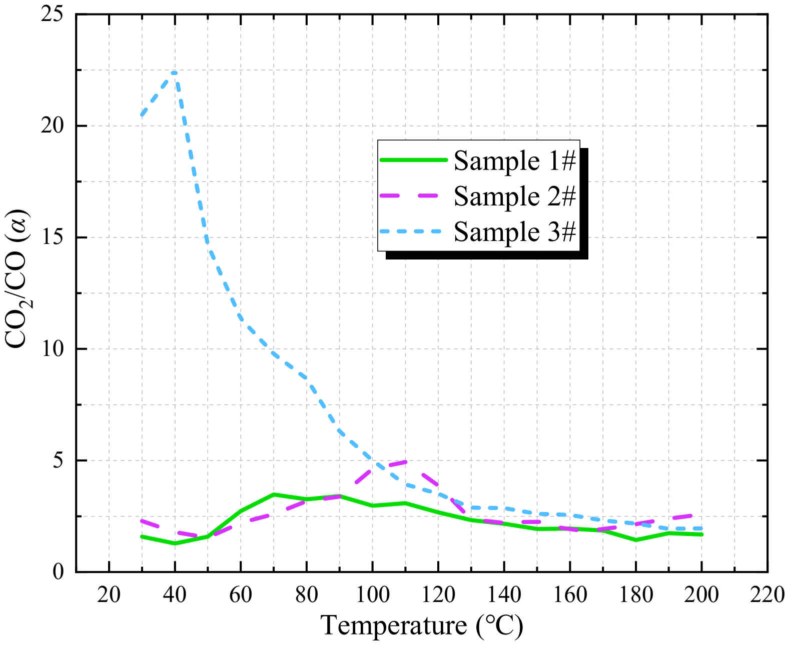

Figure 6. Variation of CO_2/CO with temperature.

3.3.2. C_2H_6/CH_4

Figure 7 demonstrates the variation of C_2H_6/CH_4 (β) with temperature. The β of lignite shows a tendency to increase and then decrease with increasing temperature, while the β of long-flame coal and poor coal generally increases with increasing temperature. Before 180 °C, the magnitude of β value is basically in the order of lignite > long-flame coal > lean coal. For lignite, after 80 °C, the production rate of C_2H_6 is greater than that of CH_4, and the β value increases rapidly, exceeding 0.3 at 100 °C. At this time, the coal sample entered the accelerated oxidation stage. Between 100 °C and 140 °C, the increase rate of β slows down, which is due to the release of large amounts of CH4 gas at high temperatures. At 140 °C, the value of β is about 0.6. After 140 °C, the production rate of C_2H_6 was lower than that of CH_4, and β showed a decreasing trend. In general, the β of lignite fluctuates greatly with temperature, and the regularity is not obvious. This could be attributed to lignite's unique coal structure and mineral composition. For long-flame coal and lean coal, the temperature point at which β begins to increase is different, 90 °C and 110 °C, respectively. They have abrupt temperature points at 130 °C and 140 °C, respectively. This means that a strong coal-oxygen complex reaction has taken place in the coal sample, and C_2H_6 gas is rapidly generated.

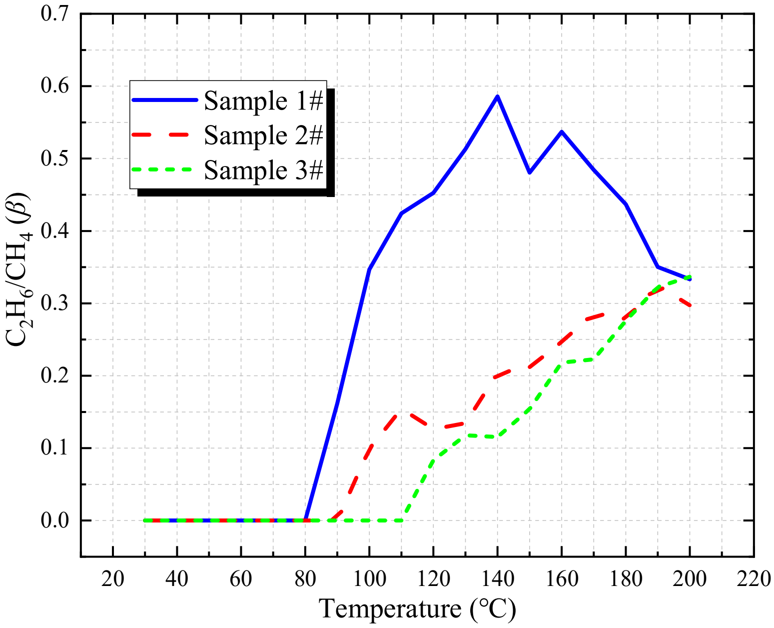

Figure 7. Variation of C_2H_6/CH_4 with temperature.

3.3.3. C_2H_4/C_2H_6 and C_2H_4/CH_4

Figure 8 demonstrates the variation of C_2H_4/C_2H_6 (γ) and C_2H_4/CH_4 (δ) with temperature. As can be seen from Figure 8a, γ shows an overall upward trend with the deepening of oxidation reaction. This indicates that the production rate of C_2H_4 is stably higher than that of C_2H_6. However, the change of γ of lignite with temperature is not obvious and fluctuates greatly. The γ of the other two coal samples basically increases with the increase in temperature. As for the long-flame coal, the temperature mutation points appeared at 120 °C, 170 °C, and 190 °C. However, the temperature mutation point of lean coal was found at 160 °C and 180 °C, respectively. In addition, it can be found from Figure 8b that δ of lignite basically shows a rapid increase and tends to be stable with temperature but still fluctuates in a small range after 130 °C. For lignite, however, a rapidly increasing mutation point occurs around 110 °C, which indicates that the coal enters a rapid oxidation stage at this stage. For both long-flame coal and lean coal, δ increases with temperature. This indicates that the production rate of C_2H_4 is stably higher than that of CH_4. Some temperature mutation points were also observed for long-flame coal and poor coal, which were 130 °C and 160 °C for long-flame coal and 160 °C and 180 °C for lean coal. In conclusion, the olefin and alkanes ratios show obvious segmentation characteristics with temperature, and they can be used as indicators to judge the spontaneous combustion process of coal to some extent.

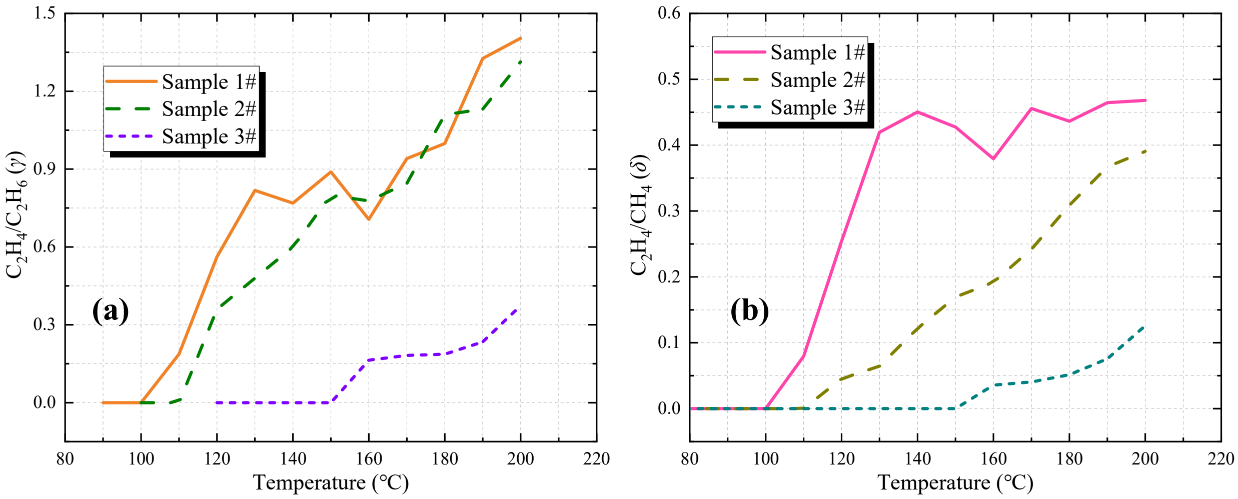

Figure 8. Changes in C_2H_4/C_2H_6 and C_2H_4/CH_4 with temperature (**a**) C_2H_4/C_2H_6 (**b**) C_2H_4/CH_4.

4. Establishment of Prediction Index System of Coal Spontaneous Combustion

4.1. Determination of Coal Spontaneous Combustion Tendency

CSC tendency is an inherent characteristic of coal spontaneous combustion, and it is the primary index for evaluating the risk of natural ignition of coal. It characterizes the possible degree of spontaneous combustion before coal mining and reflects the correlation between the physical and chemical properties of coal and its spontaneous combustion characteristics. According to the standard [31], the determination indexes of spontaneous combustion tendency of three coal samples with different coalification degrees in this paper are calculated, and their spontaneous combustion hazard grades are further evaluated. The calculation equation of the CSC tendency determination index is [31]:

$$I = \phi\left(\varphi_{C_{O_2}} \times I_{C_{O_2}} + \varphi_{T_{cpt}} \times I_{T_{cpt}}\right) - Z_1 \tag{1}$$

where, I is the determination index of coal spontaneous combustion tendency; ϕ is the amplification factor, $\phi = 40$; C_{O_2} is the oxygen concentration at the outlet of the coal sample tank when the temperature of the coal sample reaches 70 °C, %; $\varphi_{C_{O_2}}$ is the weight of low-temperature oxidation stage, $\varphi_{C_{O_2}} = 0.6$; $\varphi_{T_{cpt}}$ is the weight of the accelerated oxidation stage, $\varphi_{T_{cpt}} = 0.4$; $I_{C_{O_2}}$ is the oxygen concentration index at the outlet of the coal sample tank when the coal sample reaches 70 °C; $I_{T_{cpt}}$ is the temperature index at the intersection of coal samples under temperature-programmed condition; Z is the correction factor, Z = 300.

The calculation equation of $I_{C_{O_2}}$ is:

$$I_{C_{O_2}} = \frac{C_{O_2} - Z_{O_2}}{Z_{O_2}} \times 100 \tag{2}$$

where, Z_{O_2} is the calculation factor of oxygen concentration at the outlet of the coal sample tank, $Z_{O_2} = 15.5\%$.

The calculation equation of $I_{T_{cpt}}$ is:

$$I_{T_{cpt}} = \frac{T_{cpt} - Z_{cpt}}{Z_{cpt}} \times 100 \tag{3}$$

where, Z_{cpt} is the calculation factor of the temperature at the crossing point, $Z_{cpt} = 140$ °C.

The measured temperature at the crossing point of each coal sample and oxygen volume fraction at 70 °C were substituted into Equations (1)–(3) to calculate the determination index of each coal sample, as shown in Figure 9. After that, we obtained the spontaneous combustion propensity class of each coal sample according to the classification index of coal spontaneous combustion propensity, as shown in Table 2.

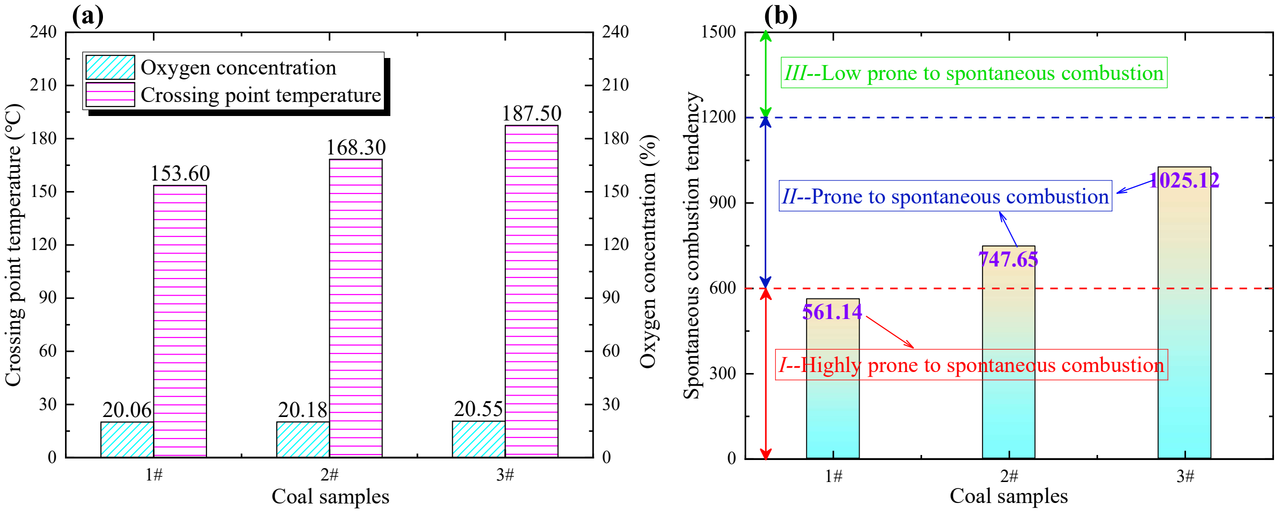

Figure 9. Spontaneous combustion tendency of each coal sample (**a**) Crossing point temperature (**b**) Determination index.

Table 2. Statistics of spontaneous combustion tendency grade and index of coal samples.

Coal Samples	Oxygen Volume Fraction at 70 °C (%)	Crossing Point Temperature (°C)	Determination Index of Spontaneous Combustion Tendency	Spontaneous Combustion Tendency Grade
Lignite	20.06	153.61	561.14	I
Long-flame coal	20.18	168.32	747.65	II
Lean coal	20.55	187.54	1025.12	II

Note: I stands for highly prone to spontaneous combustion; II indicates prone to spontaneous combustion.

From Figure 9 and Table 2, it can be concluded that the higher the coal rank is, the higher the cross point temperature and the higher the Determination index of spontaneous combustion tendency. The spontaneous combustion tendency grades of lignite, long-flame coal, and lean coal are I, II, and II, respectively, which demonstrates that these three kinds of coal samples have the danger of spontaneous combustion, and the lower the coal rank, the greater the danger of spontaneous combustion. Therefore, it is necessary to predict the process of these three kinds of coal samples to better prevent and control mine fires.

4.2. Prediction Index Optimization of Coal Spontaneous Combustion

4.2.1. Index Gases Concentration and Their Ratios

Through the analysis of the variation law of the generated gases in Section 3, it is clear that some indicator gases can be applied to predict the spontaneous combustion of coal, whereas some cannot. When selecting indicators to predict the spontaneous combustion of coal, attention should be paid to the selection of easy-to-monitor gases, and there is a good regularity between the concentration of gas indicators and coal temperature.

During the coal heating process, there is a good regularity between CO concentration and coal temperature. Generally, there will be abrupt changes at critical temperature and drying temperature. Therefore, CO concentration can be used as the main observation index to measure the stage and degree of coal spontaneous combustion. Since the emanation of C_2H_4 and C_2H_6 is unstable in the low-temperature oxidation stage, the concentration of these two gases shows an increasing trend when the temperature exceeds 120 °C, which demonstrates that the macromolecules in the coal body at this stage are not stable, the concentration of C_2H_4 and C_2H_6 is not stable in the low-temperature oxidation stage. This indicates that a large amount of gas is released from the cracking of macromolecular branch chains in coal at this stage, so it can be concluded that the coal temperature has exceeded the drying temperature. Therefore, C_2H_4 and C_2H_6 concentrations can be used as auxiliary gas indicators. Considering that some CH_4 and CO_2 occur in coal, the content of CH_4 and CO_2 varies greatly in coal under different environments and coalification degrees; it is sometimes impossible to determine whether the changing trend of CH_4 and CO_2 is caused by oxidation reaction or desorption effect [9,24,32]. Therefore, both gases are somehow not good enough to predict the spontaneous combustion of coal.

The overall CO_2/CO, C_2H_6/CH_4, C_2H_4/C_2H_6, and C_2H_4/CH_4 show obvious phasing characteristics with temperature, and some characteristic temperature points appear. It is possible to determine whether the coal has entered the stage of dry cracking and accelerated oxidation based on their trends. However, the oxidative decomposition of coal produces alkanes and olefin gases, and their generation mechanism is not clear at present. In the low temperature oxidation stage, alkanes and olefin gases are not stable and cannot react correctly with the coal temperature. In general, the ratio of alkane to olefin gas concentration can only predict the process of coal spontaneous combustion at high temperatures. Therefore, CO_2/CO, C_2H_6/CH_4, C_2H_4/C_2H_6, and C_2H_4/CH_4 can be used as indicators to assist in judging the state and degree of coal spontaneous combustion oxidation [17].

4.2.2. Fire Coefficient Index

Single gas prediction index is affected by airflow, gas extraction location, and so on, which results in misjudgment of the spontaneous combustion state of coal. For this reason, Graham put forward the concept of the fire coefficient in 1914. The percentage of fire coefficient can be calculated by the increment of CO_2 concentration ($+\Delta CO_2$), CO concentration increment ($+\Delta CO$), and reduction of O_2 concentration ($-\Delta O_2$), including the first fire coefficient (R_1), the second fire coefficient (R_2), and the third fire coefficient (R_3). The calculation formula of the specific fire coefficient is [14,33,34]:

$$R_1 = +\Delta CO_2/(-\Delta O_2) \times 100\% \tag{4}$$

$$R_2 = +\Delta CO/(-\Delta O_2) \times 100\% \tag{5}$$

$$R_3 = +\Delta CO/(+\Delta CO_2) \times 100\% \tag{6}$$

Combining with Equations (4)–(6) to analyze the data measured in the test, the three fire coefficients of the coal samples with three different degrees of coalification were calculated separately, as shown in Figure 10.

From Figure 10a, it can be clearly seen that at the initial stage of heating up, the R_1 value increases with the increase in temperature and then reaches a peak value. The peak values of different coal samples correspond to different temperature points. The corresponding temperature points of lignite, long-flame coal, and lean coal are about 60 °C, 70 °C, and 70 °C, respectively. After these temperature points, the rate of oxygen consumption is greater than the rate of CO_2 production under the faster coal-oxygen complexation, resulting in a decrease in the R_1 value. However, at around 120~130 °C, the value of R_1 begins to increase sharply. This is because the coal enters a rapid oxidation phase where the rate of increase in CO_2 concentration is greater than the decrease rate of O_2 concentration.

As can be observed from Figure 10b, the value of R_2 between 30 °C and 140 °C is relatively small, basically at about 10%. Taking lignite as an example, the R_2 value exceeds 10% at about 60 °C, which means that the coal temperature has exceeded the critical temperature and entered the stage of spontaneous combustion. Between 140 °C and 150 °C, the R_2 value exceeds 25%, and the coal-oxygen recombination is strong and enters the dry cracking stage. When the R_2 value exceeds 50% between 170 and 180 °C, it means that the coal sample has undergone a violent oxidation reaction, and corresponding countermeasures should be taken. However, long-flame coal and lean coal show a similar pattern with lignite, only the corresponding temperature turning point is different.

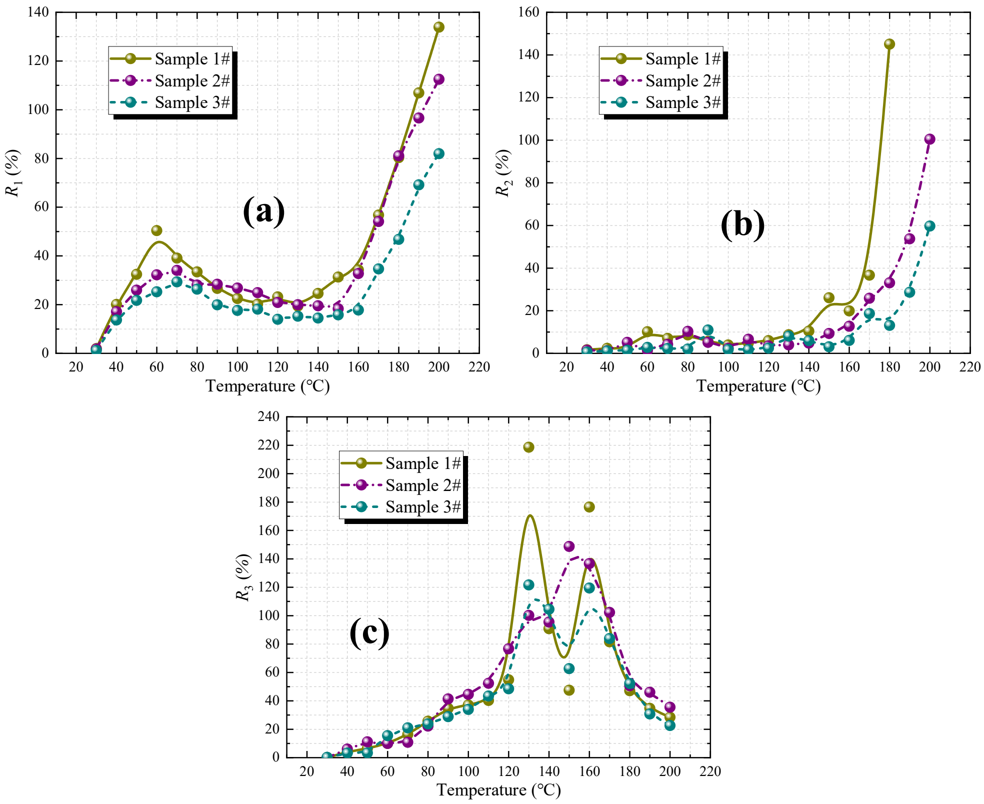

Figure 10. Variation curves of fire coefficient of different coal samples with temperature (**a**) R_1 (**b**) R_2 (**c**) R_3.

Furthermore, from Figure 10c, it can be found that the R_3 value of lignite gradually increases with the increase in coal temperature from 30 °C to about 110 °C. After 110 °C, the value of R_3 increases rapidly and reaches the first peak value. It indicates that the combination of coal and oxygen increases, and the coal enters the stage of dry cracking. After that, a peak value of R_3 appeared at 160 °C and exceeded 100%. The fluctuation of R_3 with temperature is large, and there is no obvious pattern, so it is appropriate to use the R_3 value as an auxiliary indicator. Overall, fire coefficient R_2 can comprehensively predict the entire oxidation stage of coal from low temperature to high temperature, and it is suitable to be used as the main indicator to predict coal spontaneous combustion, while R_1 and R_3 are used as auxiliary indicators.

4.3. Establishment of Prediction Index System of Coal Spontaneous Combustion

Based on the above analysis, a multi-index system can be developed to predict the spontaneous combustion state of coal, which is shown in Table 3. CO concentration, C_2H_6/CH_4, and the second fire coefficient R_2 are the main gas indexes for predicting the spontaneous combustion of coal. C_2H_4 concentration, C_2H_6 concentration, CO_2/CO, C_2H_6/CH_4, C_2H_4/C_2H_6, C_2H_4/CH_4, the first fire coefficient R_1, and the third fire coefficient R_3 were used as auxiliary gas indexes. When applying this index system, the spontaneous combustion of coal should be evaluated according to the parameters measured in the test and combined with the actual measurement in the field. During this period, relevant gas index parameters should be constantly modified to match the actual coal seam ignition situation and finally effectively guide the coal mine fire prevention and control work.

Table 3. Optimization of multi-parameter indexes for coal spontaneous combustion prediction.

Prediction Indicators	**Gas Concentration**			**Gas Concentration Ratio**					**Fire Coefficient**		
	CO	**C_2H_4**	**C_2H_6**	**C_2H_6/CH_4**	**CO_2/CO**	**C_2H_6/CH_4**	**C_2H_4/C_2H_6**	**C_2H_4/CH_4**	**R_2**	**R_1**	**R_3**
Main indicators	Yes	No	No	Yes	No	No	No	No	Yes	No	No
Auxiliary indicators	No	Yes	Yes	No	Yes	Yes	Yes	Yes	No	Yes	Yes

5. Conclusions

(i) During the heating process of the coal sample, with the increase in temperature, the oxygen concentration at the outlet decreases gradually, while the oxygen consumption rate and the concentration of each generated gas gradually increase. Oxygen consumption rate is negatively correlated with coal rank. The higher the coalification degree of coal, the more lagging some characteristic temperature points in the spontaneous combustion process.

(ii) Determination indexes of the spontaneous combustion tendency of three coal samples with different coalification degrees were determined, and their spontaneous combustion hazard levels were evaluated. Lignite is classified as Class I susceptible to spontaneous combustion, while long-flame coal and lean coal are classified as Class II capable of spontaneous combustion. Three kinds of fire coefficients were calculated to determine spontaneous combustion of coal. Taking lignite as an example, the range of critical temperature points in the spontaneous combustion process evaluated by the second fire coefficient R_2 is 60~70 °C, and the range of dry cracking temperature points is 140~150 °C.

(iii) A multi-parameter index system was established to predict the spontaneous combustion state of coal, in which the main indexes include CO concentration, C_2H_6/CH_4, and the second fire coefficient R_2. Auxiliary indicators include: C_2H_4 concentration, C_2H_6 concentration, CO_2/CO, C_2H_6/CH_4, C_2H_4/C_2H_6, C_2H_4/CH_4, the first fire coefficient R_1 and the third fire coefficient R_3. In the actual situation, continuous correction of the relevant gas index parameters is required to comprehensively determine the spontaneous combustion of coal and provide support for fire prevention and suppression work in coal mines.

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