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Study of Natural Ignition Index Gas in No. 3 Coal Seam of Yuxi Coal Mine

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Abstract To proactively mitigate the risk of coal mine fires, conduct predictive analyses before the spontaneous ignition of coal seams, ensuring the safety of coal mining operations. This study focuses on investigating the index gas associated with the natural ignition of the No. 3 coal seam at Yuxi coal mine. Temperature-programmed oxidation experiments were employed to determine the characteristic temperatures of natural ignition in the No. 3 coal seam at Yuxi coal mine. The findings indicate that the critical temperature range for the No. 3 coal seam at Yuxi coal ine is 110-130 ℃, while the dry cracking temperature range is 170-190 ℃. In identifying the index gas for the natural ignition of coal in the No. 3 coal seam at Yuxi coal mine, CO is employed, supplemented by C_2H_4, CO/CO_2, and C_2H_6/CH_4 to comprehensively assess the spontaneous combustion risk of the coal seam. This approach aims to offer a theoretical foundation and technical support for the early prediction of coal natural ignition, as well as the formulation of fire prevention and extinguishing measures for the No. 3 coal seam at Yuxi coal mine. Consequently, it provides a basis for early prediction, forecasting, and the development of fire prevention and extinguishing strategies in Yuxi coal mine's No. 3 coal seam.

Keywords Spontaneous detonation, Temperature programmed oxidation, Index gases, Forecast and prediction

1. Introduction

Coal is the main source of energy in China, with a consumption of 4 billion tons in 2021, most of which was used for thermal power generation [1, 2]. The conditions of coal beds and geological structure in China are complicated and diverse, and disasters and accidents occur frequently, among which the fire problem caused by spontaneous combustion of coal is particularly prominent[3]. Nowadays, coal mining still faces challenges from many natural disasters, and the coal spontaneous combustion (CSC) is one of the most harmful[4, 5]. Fires caused by spontaneous combustion of coal seams account for more than 90% of the total number of mine fires each year[6, 7]. In China, 56% of coal mines are threatened by the CSC [8, 9]. Therefore, effective preventive measures are the fundamental way to eliminate such accidents.

The commonly used method for predicting coal spontaneous combustion is mainly the gas analysis method, by monitoring the composition and concentration of the gas produced during coal warming and finding the variation relationship between it and the coal temperature, so as to indirectly predict the actual temperature[10-14]. Different stages of coal warming and oxidation produce different concentrations of gas components, and there is a relatively complex relationship between the concentration of these signature gases[15, 16]. For the method of gaseous forecasting, according to the physical and chemical reactions of Accepted Manuscript3 coal low-temperature oxidation, the concentration of gaseous products, such as CO, CO_2, CH_4, C_2H_4, etc., during the process of coal self-heating, it had been extensively studied[17, 18]. Meanwhile, some ratios, such as CO/ΔO_2, CO_2/CO, O_2/N_2, CH_4/C_2H_6, etc. were also recommended as forecasting indicators to evaluate the risk of coal spontaneous combustion[19, 20].

In summary, prior research on the natural ignition of coal seams has predominantly concentrated on those exhibiting a low degree of metamorphism. Anthracite coal, characterized by a high degree of metamorphism, has received less attention, attributed to its perceived resistance to spontaneous combustion. Nevertheless, in practical production scenarios, instances of natural ignition have been observed in the Yuxi coal seam at Jincheng anthracite mine. Moreover, elevated levels of CO, surpassing established limits, are recurrent in the extraction pipeline of Yuxi coal mine. To ensure the secure operation of the coal mine, optimization of the natural ignition index gas was implemented specifically for the No.3 coal seam at Yuxi coal mine. This optimization aimed to proactively prevent the spontaneous combustion of the coal seam.

2. Experimental content

2.1 Experimental principle

Spontaneous combustion of coal arises from both physical and chemical transformations within the coal, leading to the release of heat. This accumulated heat elevates the temperature of the coal body. [21]. The theory posits a coal-oxygen complex action where the dispersed coal body adsorbs oxygen, generating heat. This heat is then utilized for water evaporation, gas release, and heating of the coal body, thereby facilitating the continuation of the reaction. The ultimate goal is to reach the ignition temperature, leading to the spontaneous combustion of coal seams[22]. The oxidative heating of coal is influenced not only by the ventilation conditions within the coal mine but also by the heat dissipation processes occurring in the mine[23]. The conditions in the underground coal mine are intricate, and replicating the exact circumstances during experiments is unfeasible. Consequently, heat dissipation was disregarded in the experiments, assuming that all heat resulting from oxygen adsorption by the coal body contributed exclusively to heating the coal body. Under adiabatic conditions, oxidation and heating experiments were conducted on extracted coal samples. This facilitated the acquisition of diverse gases and concentration parameters at various stages of the coal sample oxidation process. Subsequently, an analysis of the indicative gases for the natural ignition of coal seams was carried out.

2.2 Experimental Coal Sample Analysis

The experimental coal samples were extracted from the 1304 measure lane of the 1304 working face in the No.3 coal seam of Yuxi coal mine. Subsequent to sampling, these samples underwent both industrial and elemental analyses. The outcomes of these analyses are presented in Table 1.

Table 1: Industrial and elemental analysis of experimental coal samples

Industrial analysis（%）			Elemental Analysis（%，daf）				Coal species
M_{ad}	A_d	V_{daf}	C	H	O	N	
2.37	14.28	8.07	91.19	3.24	2.27	1.31	WY3

Note: M_{ad}: Moisture (air-dry basis); A_d: Ash (air-dry basis); V_{daf} volatile matter (dry ash-free basis); daf: dry ash-free basis.

As the degree of coal metamorphism increases, so does the elemental carbon content in coal. In the coal samples from the No.3 coal seam of Yuxi coal mine, the carbon (C) content is measured at 91.19%, signifying a higher degree of metamorphism in the No.3 coal seam of Yuxi coal mine. Additionally, the hydrogen (H) content in coal seam #3 of Yuxi coal mine is recorded at 3.24%. The lower H content indicates that the coal seam in Yuxi coal mine is relatively old, belonging to a high degree of metamorphism, and is less prone to oxidation. The volatile fraction serves as the primary indicator of coal degradation, while ash represents the non-combustible mineral residue left after coal combustion. In the case of coal samples obtained from the No. 3 coal seam of Yuxi coal mine, the volatile fraction is measured at 8.07%, falling within the range of 6.5 to 10%. Additionally, the hydrogen (H) element content is recorded at 3.24%. The No. 3 coal seam of Yuxi coal mine is classified as anthracite III. This classification places the coal seam in category III for spontaneous combustion tendency, suggesting a reduced likelihood of spontaneous combustion and oxidation reactions.

The experimental coal samples were obtained from the No.3 coal seam of Yuxi coal mine, specifically from the 1304 comprehensive mining face and the 1304 measure lane. Prior to the experiment, the oxidized surface layer of the collected coal samples was removed to mitigate potential experimental errors. Subsequently, the samples underwent crushing post-sampling. To eliminate the impact of coal particle size on oxygen consumption, the experimental coal samples were processed into particles ranging from 0.15 mm to 0.25 mm, with each sample weighing 100 g. The screened coal samples were then placed in a vacuum drying box and dried at 40℃ for 48 hours to ensure that moisture did not influence the coal samples during subsequent oxidative heating.

2.3 Experimental equipment

The temperature-programmed oxidation experiment utilizes a test system designed for studying the spontaneous combustion characteristics of deep-mined coal. This experiment observes alterations in gas concentrations and temperature throughout the oxidative heating process of coal samples, employing a predetermined heating rate. Figure 1 illustrates the system configuration, comprising sample chamber, heating jacket, constant speed pump, pneumatic valve, program heating control system, gas collection system, data acquisition system, and gas analysis system.

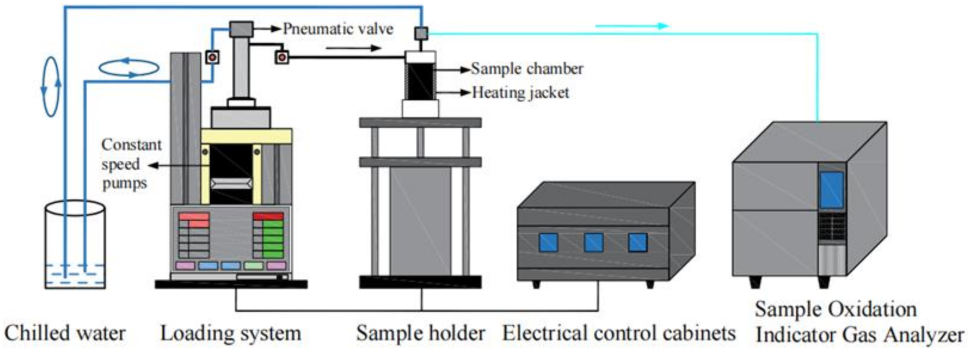

Figure 1: Test system for oxidized spontaneous combustion characteristics of deep mining coal

During the experimental process, the gas supply flow rate is set at 150 mL/min, with a gas supply oxygen concentration of 20.90%±0.5% (air). The heating rate is maintained at 1 ℃/min. Throughout the experiment, the temperature increases every 20 ℃ for gas sample collection and analysis. Each temperature block reaches the programmed value, and the holding time is set at ≥5 min to facilitate gas analysis. The objective is to measure the quantities of CO, CO_2, and hydrocarbon gases produced during the heating of coal samples. The study focuses on analyzing the composition of gas products and the variations in their concentration with temperature during the heating and oxidation of coal. This analysis also encompasses studying the oxygen consumption and exothermic characteristics of the coal samples. The goal is to determine the characteristic temperature of natural ignition for the coal samples and, ideally, identify the indicative gas for the natural ignition of the No. 3 seam in the Yuxi coal mine.

2.4 Experimental procedure

Commence by weighing 100 g of the coal sample, loading it into the sample chamber, sealing it, and verifying its gas tightness. Subsequently, initiate the coal sample heating control program. Throughout the heating process, employ the gas collection system to gather gas at preset constant temperature intervals for each temperature point. Once gas collection is concluded, utilize the index gas analyser to assess the composition and concentration of the collected gases. After gas collection, the gas analyser is employed to scrutinize the composition and concentration of the gases. Concurrently, the temperature control program records the temperature variations of the inlet gas and coal samples. The index gas analyzer is deployed to document the composition and concentration of the various gases throughout the experiment. The experiment concludes when the temperature of the coal sample reaches 200 ℃. The gas products and their concentration changes during the heating and oxidizing process of coal samples are outlined in Table 2.

Table 2: Gas concentration and coal sample temperature during heating and oxidation of coal samples

Coal sample temperature（℃）	O_2 (%)	CO (ppm)	CO_2 (ppm)	CH_4 (ppm)	C_2H_2 (ppm)	C_2H_4 (ppm)	C_2H_6 (ppm)
30℃	21.72	0	2337	1325	0	0	35.3
50℃	19.43	0.65	1435	723	0	0	36.8
70℃	19.58	20.2	3060	611	0	0	64.8
90℃	19.38	100.6	5447	267	0	0	69.0
110℃	19.43	230.8	4788	101	0	0	68.4
130℃	18.95	465.5	2529.2	59.9	0	0	46.8
150℃	17.96	1140.9	3161	66.9	0	0	21.4
170℃	16.40	3104	6442	80.2	0	0.69	7.5
190℃	10.96	8240	15341	98.99	0	2.49	2.6
200℃	5.95	12921	25817	90.16	0	4.72	1.3

The outcomes of the programmed heating experiments reveal that within the temperature range of 110-130 ℃ for the coal body, there is a noteworthy surge in the rate of oxygen consumption. This observation leads to the conclusion that the critical temperature range for the No.3 coal seam in Yuxi coal mine is 110-130 ℃. Furthermore, by examining the increasing trend in the rate of various gas productions as the temperature escalates to the range of 170-190 ℃, it is inferred that the temperature range for dry cracking is presumed to be 170-190 ℃.

3.Analysis of the pattern of generation of indicator gases

The gases produced during underground coal mine fires, induced by the spontaneous combustion of coal seams, primarily consist of CO and hydrocarbons such as CH_4, C_2H_4, and C_3H_6. By conducting experiments to simulate the process of coal heating and oxidation, this study aims to quantitatively determine characteristic parameters, including the concentration and production rate of CO, CO_2, CH_4, and other generated gases during the warming process of coal samples under controlled environmental conditions. The obtained data are used to analyze parameters like the critical temperature and accelerated oxidation temperature of the coal samples. These analyses, based on experimental results, enable the assessment of the likelihood and extent of spontaneous combustion in the coal seams.

3.1 CO generation production pattern

Figure 2 illustrates the variation in CO concentration with the temperature of the coal body for the coal sample from the 1304 working face. CO gas became detectable at 50 ℃, with a concentration of 0.65 ppm. Prior to reaching 110 ℃, the CO concentration exhibited gradual increments; however, beyond 110 ℃, the concentration of CO displayed an exponential rise in correlation with the temperature of the coal body..

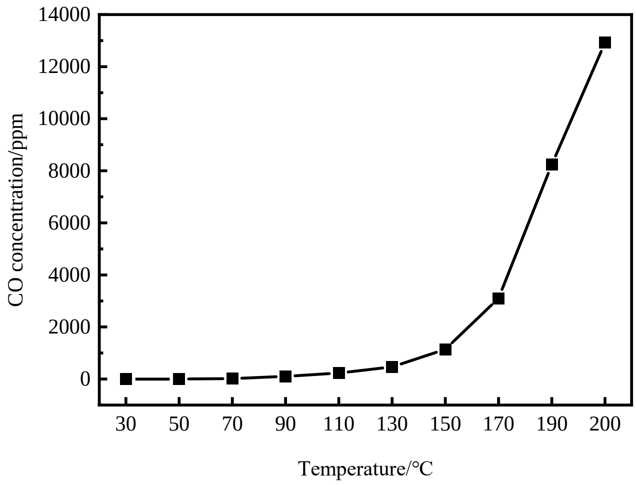

Figure 2: Variation curve of CO concentration with temperature of coal samples in 1304 working face

At 150 ℃, an anomalous spike in CO concentration occurred, signaling the onset of intense oxidation. Subsequently, CO concentration increased rapidly, marking the transition of the coal samples into a phase of vigorous oxidation reaction. The observed CO gas generation pattern during the temperature-programmed oxidation experiment aptly mirrors the characteristics of the spontaneous combustion process in the coal samples. This pattern can serve as an indicative marker gas for natural ignition in the No.3 coal seam of Yuxi coal mine.

3.2 C_xH_y gas production pattern

C_2H_4 and C_2H_2

Figure 3 depicts the variation curve of C_2H_4 concentration with the temperature of the coal body during the experimental heating and oxidation of coal samples. Analysis of Figure 3 reveals that C_2H_4 gas becomes detectable at a temperature of 150 ℃, and its concentration exhibits an approximate exponential increase with the temperature of the coal body. Notably, the concentration of C_2H_4 remains relatively small, reaching only 4.72 ppm even when the medium temperature increases to 200 ℃. Consequently, C_2H_4 can be considered as an auxiliary indicator of natural ignition in the No. 3 seam of the Yuxi coal mine. Its presence in the collected gas samples signifies that the temperature of the coal body has surpassed 150 ℃, and the coal body has entered the phase of accelerated oxidation reaction.

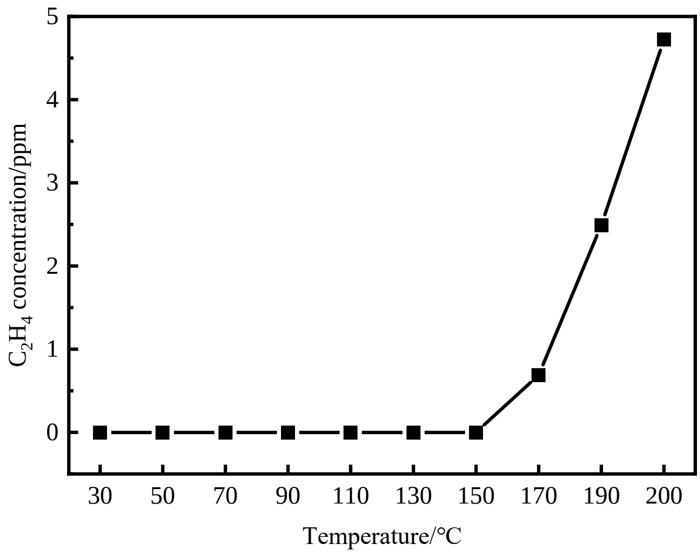

Figure 3: Variation curve of C_2H_4 concentration with temperature of coal samples in 1304 working face

Upon the onset of vigorous oxidation in the coal body, the produced gas will contain C_2H_2, which serves as an indicator reflecting the temperature range of the coal body. Despite the absence of C_2H_2 in the programmed heating and oxidation experiments, its detection in underground gas signifies that the temperature of the coal body has surpassed 200 °C, indicating that the coal body has undergone an intense chemical reaction. Consequently, C_2H_2 can be employed as one of the marker gases for the stage of vigorous oxidation in the No. 3 coal seam of Yuxi coal mine.

C_2H_6 gas production pattern

Figure 4 illustrates the pattern of C_2H_6 gas generation during the oxidation process of experimental coal samples at elevated temperatures. Analysis of Figure 4 reveals that as the temperature of the coal body reaches 30 °C, the concentration of C_2H_6 gas is measured at 35.3 ppm. Prior to the coal body temperature reaching 110 °C, the concentration of C_2H_6 gas increases in tandem with the rise in coal temperature. However, once the coal body temperature surpasses 110 °C, the concentration of C_2H_6 gas decreases with further increases in coal temperature. Ultimately, the concentration of C_2H_6 gas tends toward 0. Consequently, the fluctuation in C_2H_6 gas concentration can be utilized to ascertain the temperature of the coal body, enabling the determination of the natural developmental stage. C_2H_6 gas, therefore, serves as an auxiliary indicator for the natural ignition of the No. 3 coal seam in Yuxi coal mine.

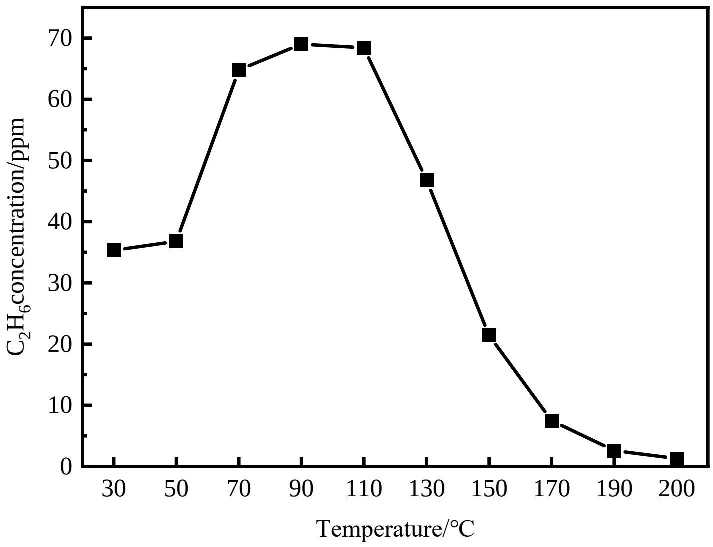

Figure 4: Variation curve of C_2H_6 concentration with temperature of coal samples in 1304 working face

3.3 Gas Ratio Analysis Streptane ratio

The chain-alkane ratio denotes the concentration ratio of a specific long-chain alkane component to the concentration of methane or ethane in the fire gas, falling within the C_1 to C_4 range. Based on the outcomes of heating and oxidation experiments conducted on coal samples from the No. 3 coal seam of Yuxi coal mine, the C_2H_6/CH_4 ratio was chosen for analysis to assess its correlation with coal's natural ignition. As illustrated in Figure 5, the C_2H_6/CH_4 ratio exhibits an initial increase followed by a decrease with the elevation of coal temperature. When the coal body temperature reaches 130 ℃, the C_2H_6/CH_4 ratio attains its peak value of 0.78. This maximum value can be employed to infer the temperature of the coal body in practical applications, allowing for the determination of the coal body's stage of natural ignition by observing changes in the C_2H_6/CH_4 ratio in collected gas samples. Thus, the C_2H_6/CH_4 ratio serves as an indicative gas for the natural ignition of the No.3 coal seam in Yuxi coal mine.

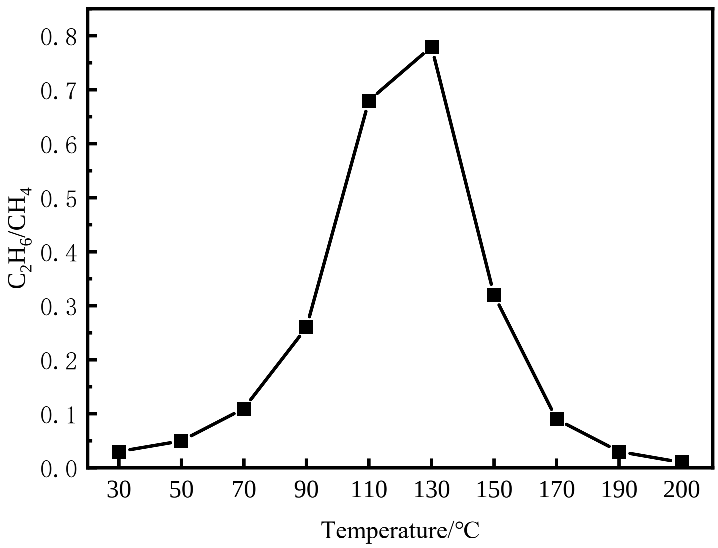

Figure 5: Variation curve of C_2H_6/CH_4 ratio with temperature

Alkane ratio

The alkane ratio denotes the concentration ratio of olefin gas to alkanes with a carbon chain equal to or greater than that of the olefin in the fire gas. Based on the outcomes of temperature programmed oxidation experiments conducted on coal samples from the No. 3 coal seam of Yuxi coal mine, the C_2H_4/C_2H_6 ratio was chosen for analysis to assess its correlation with coal's natural ignition. As depicted in Figure 6, C_2H_4 gas was not detected in the gas produced by the temperature programmed oxidation experiment before the coal body temperature reached 150 °C, resulting in a zero C_2H_4/C_2H_6 ratio. Once the coal body temperature exceeded 150 ℃, C_2H_4 gas emerged, and the C_2H_4/C_2H_6 ratio exhibited an exponential gradual increase. However, the C_2H_4/C_2H_6 ratio remained consistently below 0.1, making it prone to errors in practical applications and unsuitable for use as a marker gas for the natural ignition of the No. 3 coal seam in Yuxi coal mine.

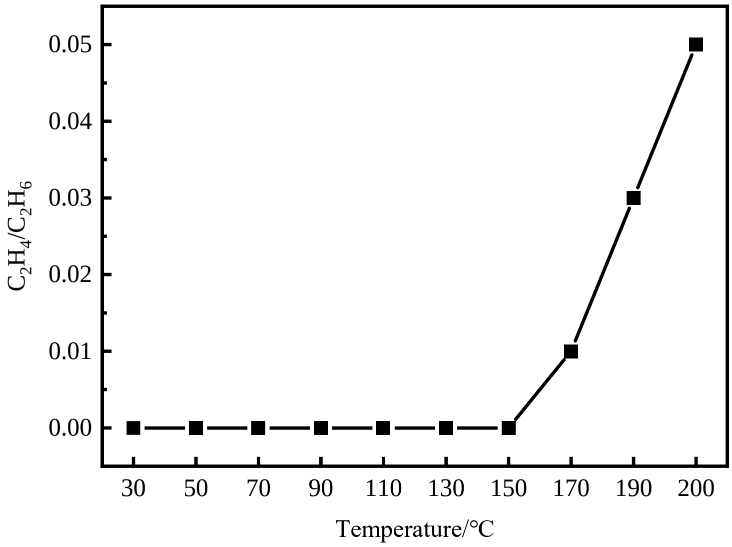

Figure 6: Variation curve of C2H4/C2H6 ratio with temperature

CO /CO_2 concentration ratio

CO serves as a sensitive indicator gas throughout the natural coal ignition process, particularly at elevated concentrations. Given the intricate underground conditions, the detection of CO may not necessarily align with the actual gas generation site. Moreover, CO is susceptible to dilution by seepage air, and its concentration is influenced by wind flow. Consequently, relying solely on CO concentration makes it challenging to assess the spontaneous combustion of a coal body. Employing the CO/CO_2 ratio to evaluate the status of coal spontaneous combustion mitigates the impact of wind flow size on gas concentration, enhancing the accuracy of judgment.

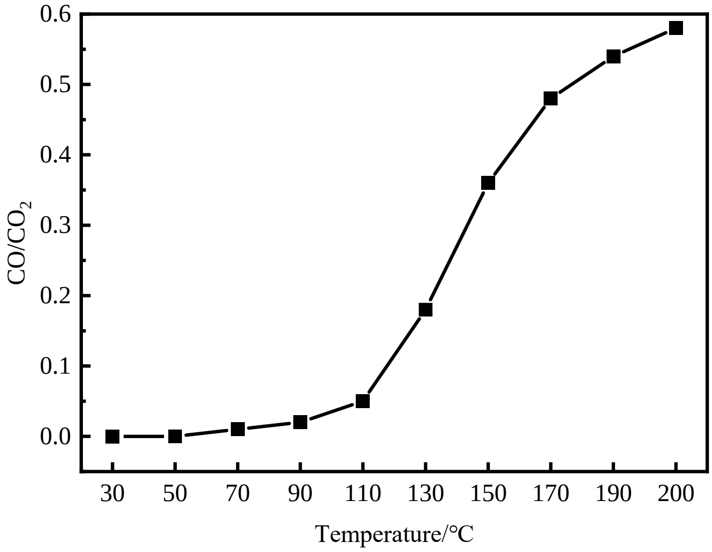

Figure 7: Variation curve of CO/CO_2 ratio with temperature

Figure 7 illustrates the variation in the CO/CO_2 ratio with the temperature of the coal body during the experimental process of coal samples. In the low-temperature oxidation stage, the CO/CO_2 ratio remains small, reaching 0.05 at 110 ℃. As the accelerated oxidation stage commences, the CO/CO_2 ratio experiences an increase, reaching 0.36 at 150 ℃ and 0.58 at 200 ℃. The CO/CO_2 ratio curve exhibits a discernible regularity, somewhat reflecting the natural ignition trend in the No.3 coal seam of Yuxi coal mine. Consequently, the CO/CO_2 ratio can serve as an auxiliary index for the identification of natural ignition marker gases in the No.3 coal seam.

4. Conclusion

1) The rate of oxygen consumption of the coal samples increases with the increase of the temperature of the coal body during the program warming process. Before 110-130 ℃, the oxygen consumption rate increases slowly with a linear law, and it can be concluded that the critical temperature range of Yuxi coal mine No.3 coal seam is 110-130 ℃; based on the growth trend of various types of gases, it is presumed that the dry cracking temperature of Yuxi coal mine No.3 coal seam is located in the range of 170-190 ℃.
2) CO was used as a marker gas in the No. 3 coal seam of Yuxi coal mine, and supplemented with C_2H_4, CO/CO_2, and C_2H_6/CH_4 to predict the spontaneous combustion of coal from Yuxi coal mine. The concentration of C_2H_4 during the experiments were small and susceptible to wind flow. Once detected underground, it indicated that the temperature of the coal body had exceeded 150 °C, indicating that the coal body had entered the accelerated oxidation stage.
3) C_2H_2 is the product of coal entering the stage of violent oxidation.C_2H_2 appeared later and produced higher initial temperature value. There was no C_2H_2 appeared during the experiment, but once C_2H_2 was detected downhole, it indicated that the coal temperature had exceeded 200 ℃ and the coal body had entered the stage of violent oxidation.

Acknowledgments

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