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A Novel Microbial Compound Inhibitor and Its Efficacy in Preventing and Controlling Mine Fire Risks: A Case Study on Low-Rank Coal Spontaneous Combustion

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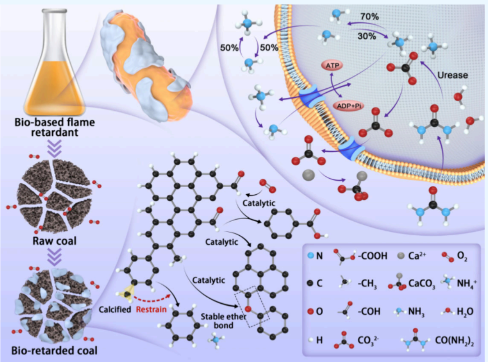
ABSTRACT: To guarantee the safety and sustainability of coal mining by effectively mitigating the substantial risk associated with coal spontaneous combustion, this study proposes a multifaceted prevention strategy aligned with green environmental principles. A compound flame retardant with a physicochemical control mechanism was prepared using indigenous microorganisms to mineralize residual coal after mining, utilizing *Bacillus pasteurelli* as a substitute material for inorganic salts. Under laboratory conditions simulating coal self-combustion, biobased flame retardants were employed to investigate the physical and chemical transformations of heat and mass evolution from ambient temperature to combustion in two representative low-rank coals. By quantitatively comparing alterations in microbiome-based groups among raw lignite, bioretarded lignite, and two control samples, the inhibitory mechanism of biobased materials on the oxygen reaction pathway was elucidated. The findings substantiated that biobased modification can consolidate the methyl and methylene groups present in aliphatic hydrocarbon side chains, which are prone to instigating low-temperature oxidation reactions. Additionally, the preventive performance of biobased flame retardants was assessed through temperature-programmed experiments, which involved estimating the critical self-heating temperature, oxygen consumption, and gas production rates of compared coal samples. The results demonstrated significant enhancements in the resistance to spontaneous combustion following bioretarded modification. Notably, the identification grade of long flame coal shifted from easy to moderate susceptibility to spontaneous combustion. Furthermore, biobased flame retardants exhibited remarkable flame retardancy rates of approximately 80% for lignite, thereby validating their efficacy as more environmentally friendly and technologically advanced substitute materials for inhibiting spontaneous combustion in low-rank coals.



1. INTRODUCTION

The spontaneous combustion of coal is a significant factor contributing to mine fires, and its combustion process is essentially a self-sustaining oxidation reaction. Understanding the mechanism of spontaneous coal combustion can enable timely intervention and prevention of the process, ensuring the safety of underground workers' lives and property. Currently, researchers in related fields have conducted extensive studies on the existing theory of coal-oxygen recombination and have further proposed the chain reaction theory based on adsorption and free radical transformation mechanisms, providing insights into the chemical perspective of organic compound molecule transformation during coal oxidation.[1−3]

Oxygen comes into contact with the surface of the coal, leading to the adsorption of active substances (such as oxygen and nitrogen oxides) on the coal surface, generating free radicals. These free radicals then react with adsorbed oxygen to release heat and produce oxidation products. If a sufficient amount of oxygen is present, then this reaction will continue until the internal temperature of the coal reaches its self-heating critical point. At this stage, biomethane, pyrolytic methane, and carbon radicals in the coal undergo initial reactions with oxygen to generate additional oxides, while rapidly elevating the temperature. Simultaneously, hydroxyl and fat free radicals participate in this chain reaction. Oxidation leads to the production of various oxidation products, such as carbon monoxide, carbon dioxide, methane, and other flammable gases. These substances play a crucial role in the chain reaction by releasing significant amounts of heat and continuously depleting the active sites of coal and oxygen. If the heat generated by the coal surpasses the heat dissipated into the surroundings, then the temperature will continue to rise, potentially resulting in smoke and open fires.

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According to existing research, the continuous process of coal spontaneous combustion requires combustible material, oxygen, and a heat source. If any of these elements are disrupted, then the combustion process can be inhibited or stopped. In light of this, various measures have been proposed by predecessors to control the spontaneous ignition. Compared to grouting, gel injection, and three phase foam fire prevention methods, traditional grouping technology experiences a significant reduction in flame retardant performance due to water loss in the slurry. To prevent the occurrence of spontaneous coal combustion before natural oxidation begins, flame retardants can be sprayed or dispersed on the surface of the coal in advance. The on-site application equipment and operation process not only are more convenient but also provide stronger durability as the flame retardant solution's chemical components continue to play a role even if the sprayed surface of coal evaporates or is lost. This flame retardant technology, commonly using inorganic salts such as $CaCl_2$, $MgCl_2$, and NaCl, effectively cuts off oxygen and provides water cooling to cool the coal body. Continuous efforts have been made to enhance the flame retardant efficiency and prolong the duration of action of flame retardants through the ongoing research and development of their composition. This has led to the development of chemical flame retardants and physical-chemical composite flame retardants, building upon the foundation of physical flame retardants that mitigate coal's propensity for spontaneous combustion from a chemical perspective. Chemical flame retardants function by disrupting the free radical chain reaction in coal through trapping active free radicals generated during oxidation reactions.[4] They enhance the activation of chemisorption during low temperature oxidation to function as a flame retardant[5] and facilitate the formation or consumption of specific functional groups within the coal body. Some findings from current research on composite flame retardants are presented in Table 1.

The aforementioned composite flame retardants used in coal mining sites must come into direct contact with the coal to effectively inhibit flames. However, during this process, the active components of the flame retardants evaporate into the surrounding soil, leading to a loss of reactivity and a potential environmental impact on soil acidity and groundwater. To address these concerns, utilizing naturally occurring organic minerals with synthetic modifications for flame retardancy will significantly enhance the environmental sustainability of fire prevention measures, in line with green mining principles. Therefore, the research and development of new composite flame retardants based on organic compounds and microorganisms that rely on organic compounds for survival and metabolism hold significant research value and promising application prospects. Building upon this concept and previous studies, this paper proposes a novel composite flame retardant utilizing MICP technology, specifically the microbe-induced calcium deposition mechanism, to effectively inhibit coal spontaneous combustion. The core product, calcium carbonate, can substantially alter the physicochemical properties of the modified materials. The application of this technology in the construction field dates back to the early 1980s, and significant progress has been made in various areas, including repairing concrete cracks, enhancing concrete strength, removing calcium ions in sewage, improving the soil environment, and preventing water seepage in dams.[24,25] Currently, the application of MICP technology in the coal mining industry is still under development. Urease bacteria can hydrolyze urea to generate carbonate ions in an environment containing urea and calcium sources. They also adsorb free calcium ions in the environment to bind coal particles and form calcium carbonate crystals that encapsulate coal particles. Based on this characteristic, some researchers have suggested the application of MICP technology to control secondary dust in coal mining.[26] They also proposed the preparation of a microbial dust suppressor for mining with the aim of improving coal dust wettability and enhancing coal dust consolidation degree, which has been proven to effectively consolidate coal dust.[27] Additionally, previous studies successfully isolated urease-producing bacteria from coal and conducted a series of dust suppression experiments using these microorganisms. This confirmed the natural occurrence of urease-producing microorganisms in coal and demonstrated the consolidating effect of their metabolites on crushed coal powder.[28,29] This discovery means that MICP technology should be applied to the research and development of new coal flame retardants. The use of the same microorganisms in coal mine flame retardants offers clear ecological advantages, as it minimizes the impact on surrounding biodiversity. Additionally, the biological components in the flame retardant material can maintain activity for an extended period under suitable environmental conditions, providing prolonged flame retardant effects when sufficient calcium sources are present. This promotes the mutual adaptation between the environment and organisms. In a urea-containing environment, urea diffuses freely into the cell interior through the membrane of urease microorganisms,[30] where it is decomposed by urease into NH_4^+ and CO_3^{2-} ions[31] and then transported outside the cell. During this process, microorganisms continuously adsorb Ca^{2+} ions in the environment as crystal nuclei. Ultimately, calcium carbonate precipitates with CO_3^{2-} ions on the surface of the cell membrane,[32] effectively blocking primary coal pores and isolating oxygen from contacting the coal surface. At the same time, the biological flame retardants can react and consume the active functional groups within the coal, catalyze the formation of stable ether bonds within the coal, and solidify a portion of the reactive side chains. This makes it difficult for them to

Table 1. Classification of Composite Flame Retardants

Flame Retardants	Material
corrosion inhibitor gels	tea polyphenols modified with polypropylene and mixed with halloysite nanotubes[6,7]
	proanthocyanidins and mixed with ammonium polyphosphate[8]
	biomass thermoresponsive konjac glucomannan[9]
	sodium-based bentonite junction and kaolin, silica, hydrogen peroxide, and silicon oxide[10]
	polyacrylamide (PAM) and the nanomaterial silica aerogel powder (SA)[7]
composite corrosion inhibitor foams	thickener and cross-linker[11,12]
	dispersed particle gel (DPG)[13]
	compound foaming agent (CFA) and high water absorbency gel (HWAG)[14]
	sodium dodecyl sulfate (SDS) and NH4Cl, $MgCl_2$[15]
	polymorphic foam (PF) synthesized from a sol foam/polycaprolactone (SFP) solution and organic acid[16]
halogen corrosion inhibitors	$MgCl_2$ + NaCl, $MgCl_2$ + $CaCl_2$[17]
	$MgCl_2$ and phosphates[18]
natural organic corrosion inhibitors	catechin and poly(ethylene glycol)[19]
	superabsorbent hydrogel−ascorbic acid composite[20]
	a chelated calcium procyanidine-attapulgite[21]
	D,L-malic acid[22]
	metal chelating agent, citric acid[23]

participate in the coal oxygen reaction, effectively slowing the oxidation process of coal, as depicted in Figure 1.

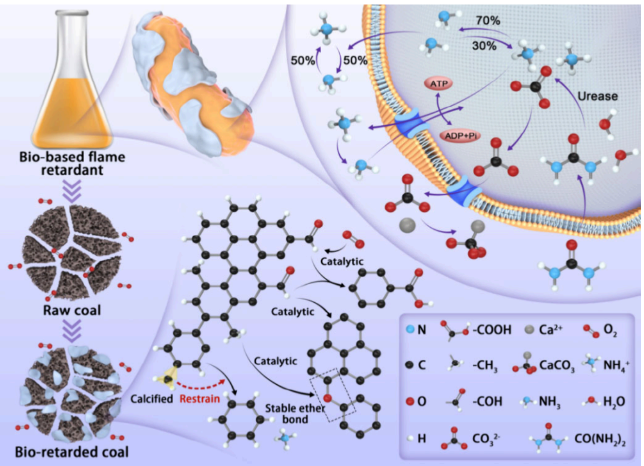

Figure 1. Schematic diagram illustrating the retarding mechanism of biobased flame retardants on coal spontaneous combustion.

Based on the aforementioned summary and analysis, *Bacillus pasteurelli* was selected as the experimental strain in this study to explore a novel type of biobased flame retardant for preventing coal spontaneous combustion; $CaCl_2$ as a traditional chemical component of flame retardant was incorporated into the biological culture medium of the *Bacillus* as well. By employing a steep ascent design (SAD) experiment and utilizing response surface methodology (RSM),[33] optimal culture conditions were determined to achieve the highest OD600 concentration of bacterial liquid. Furthermore, the ratio between bacterial solution and $CaCl_2$ reagent that resulted in maximum calcium carbonate deposition mass was also identified, leading to the successful development of a pioneering biobased flame retardant based on inorganic salt. The investigation primarily focused on the heat and mass changes observed during the spontaneous combustion process of coal samples both before and after treatment. This was done to elucidate the physical and chemical properties of bioflame retardant materials in preventing spontaneous combustion. Through synchronous thermal analysis experiments, in situ Fourier infrared spectroscopy tests, and programmed temperature experiments, we studied the variation in content of each functional group during heating processes and analyzed the intersection temperature of the coal seam during the low-temperature oxidation stage. Furthermore, by analyzing gas production and oxygen consumption rates, we quantitatively investigated the mechanism through which a novel biobased flame retardant inhibits spontaneous combustion in low-rank coal.

2. MATERIALS AND METHODS

2.1. Preparation of Experimental Reagents and Sample.

To prepare biobased flame retardants, the initial step involves obtaining a *Bacillus pasteurelli* solution through cultivation. 3 g of beef extract was combined with 5 g of peptone in a sterilized container filled with 1000 mL of distilled water. The mixture was thoroughly stirred until complete dissolution, and the container was tightly sealed before storing it at room temperature. The NaOH solution was precisely diluted to achieve a pH range between 7.5 and 8.0, and then it was evenly poured into a conical bottle while ensuring proper closure. Following high-temperature sterilization, the prepared urea solution was introduced and an alcohol lamp was ignited for inoculation using an inoculation ring. Culturing was proceeded on a constant temperature shaking table set at 30 °C and 180 rpm for a duration of 48 h.

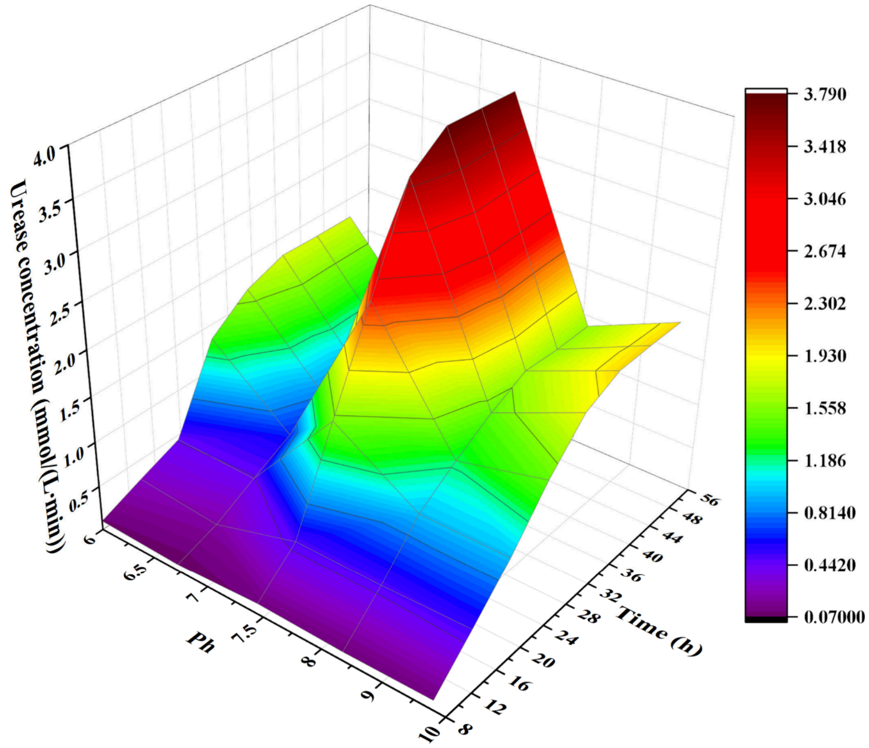

Figure 2. Response surface of the urease activity for multiple control parameters.

In the process, a higher urease activity indicates a faster decomposition rate of urea and an increased production of calcium carbonate precipitation per unit time, which directly affects the resistance of the bioflame retardants. The culture of *Bacillus pasteurelli* must consider the microbial liquid medium, temperature, pH, and other environmental conditions that impact the abundance and activity of microorganisms, subsequently influencing the formation efficiency of calcium carbonate. The OD600 measurement provides an accurate means to quantify microorganism numbers in a liquid medium by utilizing the absorption spectrum at a wavelength of 600 nm. This technique relies on monitoring real-time changes in transmittance and reflection spectra as indicators of increasing sample concentration. The OD600 value reflects the bacteria's capacity to produce urease, which directly influences the activity of urease in the culture medium. The determination method for urease activity is based on the ion balance principle of solution: when 1 mol of urea is mixed with bacterial solution, it will be decomposed by urease into 2 mol of NH_4^+ and 1 mol of CO_3^{2-}, thereby affecting its conductivity. Therefore, this change can be used to estimate the urease activity in solution, as shown in eq 1

$$U = \Delta\sigma \times 11.11 \tag{1}$$

where U is urease activity, m·mol·L^{-1}·min; $\Delta\sigma$ is change in conductivity, mS·cm^{-1}·min.

To enhance the optimization of biobased flame retardant preparation, steep ascent design (SAD) tests were conducted using Design Expert 10. The investigation revealed that key factors influencing the preparation process included culture time, temperature, pH value, and the ratio of culture solution components (concentration of cement solution, ratio of cement solution to bacterial solution, and ratio of urea to calcium source). To further refine the optimization process for biobased flame retardant preparation, a Box-Behnken model was employed to design an experiment utilizing response surface methodology (RSM). This approach aimed to delve into the interplay between these conditions and identify the optimal solutions impacting microbial mineralization processes.

2.1.1. Optimization of Bacterial Culture Medium. The optimal temperature for Bacillus enzyme activity is approximately 30 °C, which facilitates efficient biochemical reactions and metabolic processes within the cell, leading to enhanced growth. Consequently, experiments employing the response surface methodology to investigate environmental pH and culture duration as critical factors are conducted at an ambient temperature of 30 °C. The pH values were adjusted to 6, 6.5, 7, 7.5, 8, 9, and 10, in five different media. Five groups of mutual control experiments were established. A sterilized plastic head dropper was completely emptied, and a specific amount of *Bacillus pasteurelli* was absorbed and dropped into five Petri dishes individually. The culture dishes were continuously observed, recording microbial concentrations when reaching specific time intervals including 8, 16, 24, 40, 48, 56, and finally 60 h; urease activity was also calculated during these time points. The response surface method was employed to analyze the collected data with results presented in Figure 2, with the Z-axis denoting urease activity and the X- and Y-axes representing pH value of the culture environment and duration of cultivation, respectively.

2.1.2. Optimization of Compound Parameters. Calcium carbonate, as a metabolite of *Bacillus*, is directly influenced by the environmental conditions of the calcium source, thereby impacting its quality. To ascertain the optimal conditions for fostering the production of high-quality calcium carbonate by *Bacillus pasteurelli*, a sequence of controlled experiments was conducted using the biobacterial liquid prepared with the aforementioned optimal ratio. The generation of calcium carbonate was assessed and documented across varying concentrations of cementing fluid and different proportions of urea and calcium sources. The experimental findings are depicted in Figure 3. Notably, the peak yield of calcium carbonate was observed at a cementing fluid concentration of 1 M. Furthermore, adjusting the ratio of urea to calcium source under the condition of 1 M cementing fluid concentration allowed for the evaluation of calcium carbonate yield in the Petri dish. The optimal calcium carbonate yield was determined when the ratio of urea to the calcium source was 1:1.5.

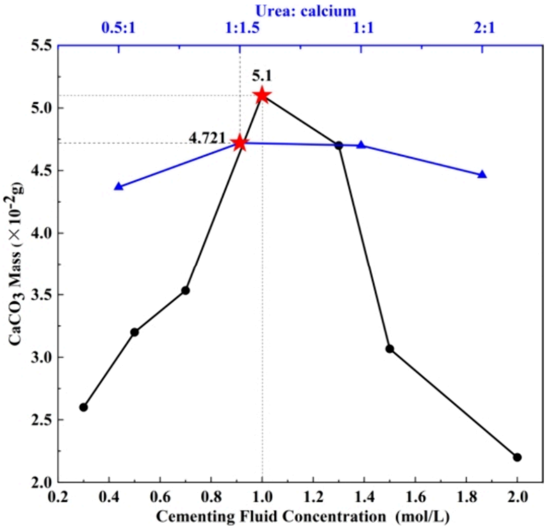

Figure 3. Calcium carbonate production under different culture conditions.

2.1.3. Preparation of Experimental Coal Samples. According to the optimized formula obtained in the previous experiment, which achieved maximum urease activity and high-quality calcium carbonate generation of the bacterial solution, the optimal range for each variable was determined. The culture temperature was set at 30 °C with a culture time of 48 h. The pH value of the culture solution was adjusted to 7.602 to prepare the bacterial solution. The concentration of the cementing solution was set at 1 M, and a ratio of 1:1 between the cement solution and bacterial solution was maintained. Under these conditions, a biobased flame retardant material can be configured by combining urea and the calcium source in a ratio of 0.667. Figure 4 illustrates the prepared biobacterial liquid.

The lignite and long-flame coal were chosen as the research subjects, and the fresh coal samples were hermetically sealed with plastic wrap for transportation to the laboratory. Following crushing treatment, the samples were divided into 40−80 mesh and 200−300 mesh sizes and then placed in a vacuum environment for 48 h to undergo drying in preparation for subsequent utilization. According to the relevant industry standards, the industrial analysis and elemental analysis results of the two coal samples were obtained.[34] The results of this analysis are presented in Table 2.

The biobased flame retardant was prepared by combining 5 mL of a calcium chloride solution with a concentration of 1 mol/L and 5 mL of a microbial culture medium (bacterial activity

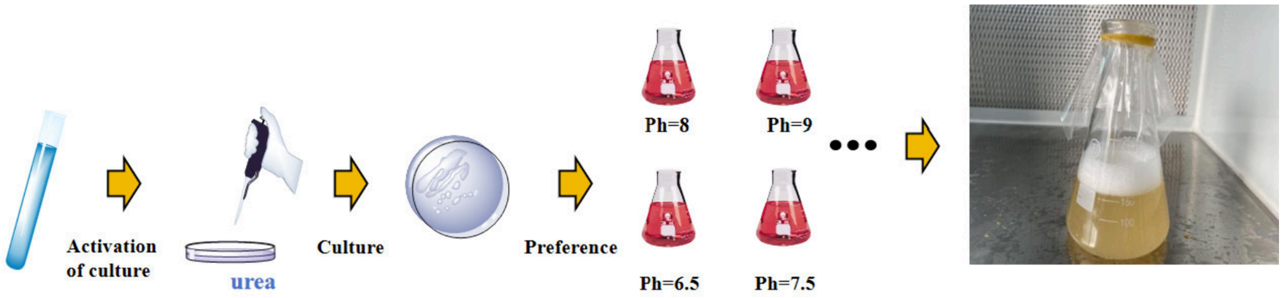

Figure 4. Optimal culture of the bacterial solution.

Table 2. Proximate and Ultimate Analyses of Coal Samples

Industrial Analysis				Elemental Analysis				
Coal Sample	Mad (%)	Aad (%)	Vad (%)	Cad (%)	H (%)	O (%)	N (%)	S (%)
lignite	0.86	6.81	23.15	69.18	4.54	29.19	0.93	0.34
long flame coal	1.58	6.92	30.71	80.97	4.30	12.49	0.97	1.27

OD600 = 1.6) that had been cultured for 48 h. The samples of biobased flame retardant lignite and biobased flame retardant long flame coal were obtained by immersing 5 g of lignite and long-flame coal powder, respectively, for a duration of 24 h. To prepare the chemical flame retardant required for the experiment, 5 mL of a sodium bicarbonate solution with a concentration of 1 mol/L was mixed with an equal amount (5 mL) of the aforementioned calcium chloride solution. 5 g of lignite coal and long-flame coal were immersed in the mixture for 24 h to prepare the chemical flame retardant coal sample. Under the same conditions, two types of coal samples were submerged in water to obtain immersed coal samples. The three sets of experimental coal samples prepared, along with untreated raw lignite and long-flame coal, were placed in a drying oven at 40 °C under vacuum for 48 h, sealed, and stored.

2.2. Experimental Methods. *2.2.1. Synchronous Thermal Analysis Experiment.* The experiment was conducted at the School of Safety Engineering, China University of Mining and Technology, utilizing TA Company's SDT-Q600 synchronous thermal analyzer. The experimental parameters encompassed a coal sample mass of 10 mg, a temperature range spanning from room temperature to 800 °C, a carrier gas flow rate set at 100 mL/min, and a heating rate maintained at 10 °C/min. Prior to commencing the experiment, heat flow and thermogravimetric curves were measured employing an empty crucible for the coal sample; subsequent results were then calculated based on the specified experimental conditions.

2.2.1.1. Thermogravimetric Experiment. The thermogravimetric (TG) analysis, along with its associated heat curves, allows for a comprehensive examination of four experimental lignite coal samples from various perspectives using a synchronous thermal analyzer. This enables precise temperature control and determination of the correlation. between temperature (T) and mass. Consequently, monitoring the temperature fluctuations of coal samples through thermocouples and thermobalances facilitates the construction of TG curves that accurately depict the actual characteristics of the samples. Additionally, drawing DTG curves provides a clear representation of how the sample mass changes in relation to increasing temperatures.

By utilization of differential scanning calorimetry (DSC), it is possible to achieve rapid heating of the experimental specimen, enabling the determination of the energy difference between the experimental and reference objects. This disparity can be quantified as a DSC curve, which effectively illustrates the temporal temperature variations in the experimental object. Within this curve, each moment exhibits varying heat levels over time, resulting in an overall increase in its area. The TG curve provides insights into mass loss during coal-oxygen composite reactions, while the DTG curve elucidates changes in weight loss rates throughout oxidation processes.[35] Additionally, the DSC curve showcases heat release trends with respect to temperature fluctuations. Through the meticulous analysis of thermogravimetric data obtained during coal oxygen heating experiments, it becomes feasible to discern six distinct characteristic temperature points that delineate different stages within this process: critical temperature T1, dry cracking temperature T2, maximum mass temperature T3, ignition point temperature T4, maximum thermogravimetric rate temperature T5, and burnout temperature T6.[36]

2.2.1.2. In Situ Fourier Infrared Spectroscopy Experiment. The experiments were carried out on a VERTEX 80v infrared spectrometer. A sample of 1 g of solid coal powder with particle size between 200 and 300 mesh was prepared. The test sample was made by mixing KBr with the coal sample and pressing. The scanning range was between 4000 and 400 cm^{-1}, and 64 sample scans were completed.

2.2.2. Analysis of Oxidation Kinetics. By employing kinetic analysis, we can gain a more comprehensive understanding of the distinct characteristics exhibited by the "kinetic three factors".[37] This enables us to delve deeper into their intricate interplay and consequently enhances our ability to accurately forecast the trajectory of coal oxidation reaction development. The low-temperature oxidation process of coal can be succinctly summarized as follows.

$$\mathrm{Air\ (g) + Coal\ (s) + Reaction\ atmosphere\ (g)} \tag{2}$$

Through the continuous supply of oxygen to the coal sample, it has been observed that the gas generated by the reaction is effectively carried away by the air flow. This observation indicates an irreversible nature of the reaction during the low temperature oxidation process. The relationship between coal mass and heat, as well as its impact on the reaction rate, manifests in two distinct forms.

Integral form:

$$G(\alpha) = k \times t \tag{3}$$

Differential form:

$$\frac{\mathrm{d}\alpha}{\mathrm{d}t} = k \times f(\alpha) \tag{4}$$

where α is the coal conversion rate at time t, k is the reaction rate constant, G is the integral form of the reaction mechanism function, and f is the differential form of the reaction mechanism function.

According to the Arrhenius equation, the relationship between k and the reaction temperature T (absolute temperature) can be expressed as

$$k = A \times \exp\left(\frac{-E}{RT}\right) \tag{5}$$

where E is the apparent activation energy of the reaction, kJ/mol; A is the apparent pre-exponential factor of the reaction, s^{-1}; R is the universal gas constant, 8.314 $\mathrm{J{\cdot}mol^{-1}{\cdot}K^{-1}}$

Equation 5 is combined with eqs 2–4 to obtain the commonly used kinetic equation of a heterogeneous system under isothermal conditions:

$$\frac{\mathrm{d}a}{\mathrm{d}T} = \frac{A}{B} \times f(\alpha) \times \exp\left(\frac{-E}{RT}\right) \tag{6}$$

where β is the heating rate, $°\mathrm{C{\cdot}min^{-1}}$, $f(\alpha) = (1 - \alpha)$.

The formula is integrated and treated with Coats-Redfern. After finishing, it can be obtained.

When $n \neq 1$,

$$\ln\left[\frac{(1-(1-\alpha)^{1-n})}{(1-n) \times T^2}\right] = \ln\left[\frac{AR}{\beta E}\left(1 - \frac{2RT}{E}\right)\right]\frac{E}{RT} \tag{7}$$

When $n = 1$,

$$\ln\left[\frac{(-\ln(1-\alpha))}{T^2}\right] = \ln\left[\frac{AR}{\beta E}\left(1 - \frac{2RT}{E}\right)\right] - \frac{E}{RT} \tag{8}$$

Equations 7 and 8 are the Coats-Redferns equations. When $n = 1$, $\ln[-(\ln(1-\alpha))/T^2]$ to $1/T$ was plotted, where the slope is $-E/R$ and the intercept is $\ln(AR/\beta E)$; then E and α can be found. When $n \neq 1$, $1/T$ with $\ln[(1-(1-\alpha)^{1-n})/(1-n)T^2]$ is plotted, also according to the intercept and slope of the line, and activation energy and frequency factor A can be obtained.[38] Therefore, no matter what the value of n is, as long as the graph of $\ln[G(\alpha)]/T^2 - 1/T$ is present, the value of E can be accurately calculated.

2.2.3. Programmed Temperature Experiment. The temperature-programmed experiment was conducted on lignite and long-flame coal test samples using the coal spontaneous combustion tendency detection system developed by the China University of Mining and Technology. The hydrogen generator was turned on; the hydrogen flow rate reached 340 mL/min. Then, the meteorological chromatograph was started, and we waited for its parameters to stabilize. The calibration of the meteorological chromatograph was carried out, including activating the hot load bridge flow and ignition, using three channels A, B, and C to detect the gas, and passing the standard gas for analysis and correction. 50 g of coal sample with particle size of 40–80 mesh was weighed. It was put into an iron coal sample tank, which was placed into an iron container. Then, it was covered with asbestos and put it into programmed temperature box. The inlet, outlet, and temperature probes were then connected. The parameters of the program heating box were set, and air was injected when the temperature reached 30 °C. The heating rate was set to 0.8 °C/min, and the gas concentration was measured every 10 °C until the temperature reached 230 °C.

At the same temperature, a lower oxygen concentration leads to increased oxygen consumption of the coal sample, resulting in a more vigorous reaction and a pronounced tendency toward spontaneous combustion. Therefore, the O_2 concentration and oxygen consumption rate at the exit are utilized as index parameters for analyzing the severity of the reaction in experimental coal samples during low-temperature stages. Experiments have demonstrated that there is a direct correlation between the increase in the temperature and the corresponding increase in the oxygen consumption of coal samples. Considering the gradual rate at which the furnace temperature increases, it can be assumed that the furnace temperature reaches a state of equilibrium distribution. Consequently, the calculation for the oxygen consumption rate per unit volume of coal sample is as follows:

$$V_{O_2}(T) = -\frac{\mathrm{d}C}{\mathrm{d}\tau} = -\frac{\mathrm{d}C}{\mathrm{d}x/\mu} = -\left(\frac{\mathrm{d}C}{\mathrm{d}x}\right) \times \left(\frac{Q}{S}\right) \tag{9}$$

where, $V_{O2}(T)$ is the oxygen consumption rate of coal body at temperature T, $\mathrm{m^3/(m^3{\cdot}s)}$; C is the oxygen concentration at the outlet of the coal sample tank, %; τ is time, s; μ is the airflow velocity in the coal sample tank, m/s; x is the distance from any point in the tank to the inlet of the coal sample tank, m; Q is the air supply volume, 1.6×10^{-6} $\mathrm{m^3/s}$; S is the cross-sectional area of the coal sample tank, 15.9×10^{-4} $\mathrm{m^2}$.

Using the oxidation kinetic analysis technology, we can calculate the comprehensive evaluation index of coal spontaneous combustion, which is expressed as follows:

$$V^T = K{\cdot}C \tag{10}$$

We can derive the standard oxygen consumption rate formula:

$$V^0_{O_2}(T) = \left(\frac{C_0}{C}\right)V_{O_2}(T) \tag{11}$$

where $V^0_{O_2}(T)$ is the oxygen consumption rate at temperature T and oxygen concentration C_0, $\mathrm{m^3/(m^3{\cdot}s)}$; C_0 is the oxygen concentration in the coal sample tank, the value of which is 20.96% in this paper. The data is substituted, and the integral on both sides is obtained:

$$V^0_{O_2} = \left(\frac{QC_0}{S \times L}\right) \times \ln\left(\frac{C_0}{C}\right) \tag{12}$$

where L is the height of the coal sample, which is 0.065 m.

3. RESULTS AND DISCUSSION

3.1. Bioretarded Mechanism Analysis.

3.1.1. Infrared Spectroscopic Analysis of Spontaneous Combustion Process. In situ infrared spectroscopy was employed to analyze the molecular structural changes of the coal samples during the heating process. Figure 5 illustrates the infrared spectra within the 400–4000 $\mathrm{cm^{-1}}$ range for both lignite raw coal and coal samples treated with two flame retardants.

According to the theories of coal-oxygen recombination and free radicals, upon contact between coal and oxygen, highly active free radicals and functional groups in coal react with oxygen to form peroxides. This reaction further generates new free radicals, small molecule hydrocarbons, and other products

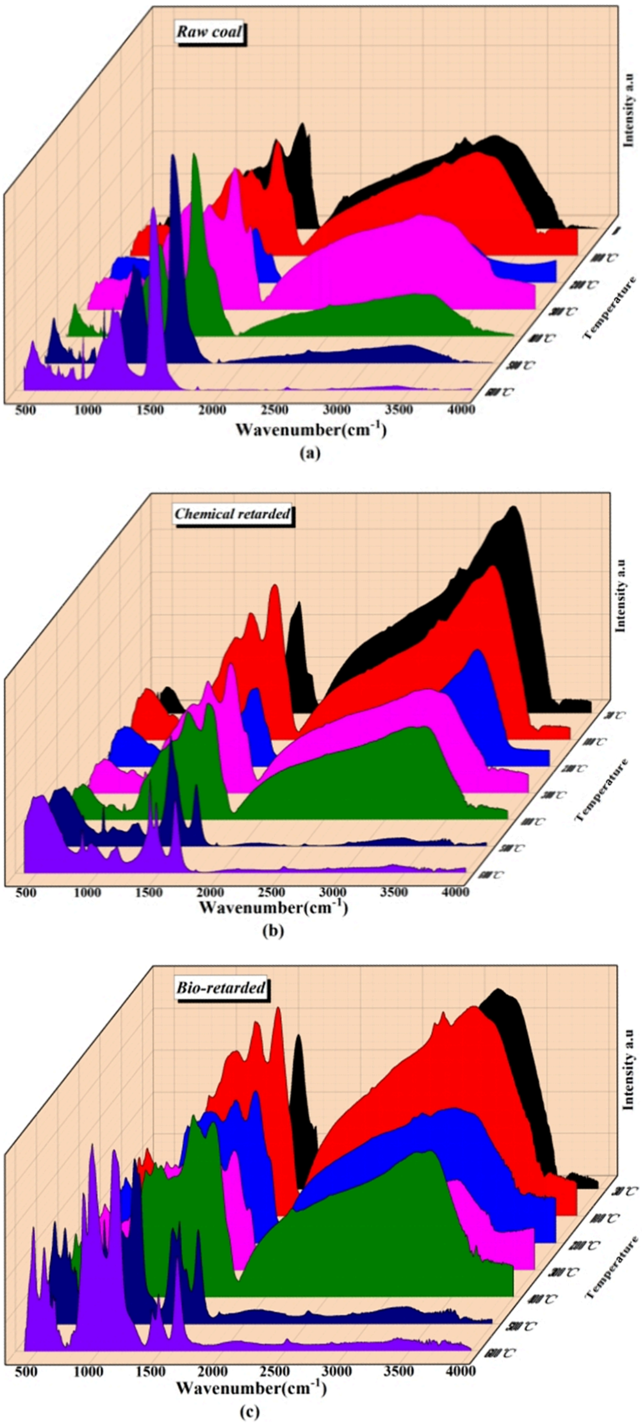

Figure 5. Infrared spectrum of lignite during spontaneous combustion process: (a) raw coal, (b) chemical retarded lignite, and (c) bioretarded lignite.

that promote chain reactions and accelerate the internal heating. Through extensive experimentation and analysis by previous researchers, the elementary reactions occurring during the oxidation stage of coal have been elucidated. It has been determined that the methyl and methylene groups on the side chains of aromatic rings are the main functional groups involved in the coal-oxygen reaction. Additionally, oxygen-containing functional groups such as hydroxyl and carboxyl groups continuously emerge as important intermediates throughout the entire oxidation process while being generated and consumed. Ultimately, these intermediates transform to relatively stable ether bonds. Figure 6 illustrates a typical initial primitive reaction observed during the spontaneous combustion of coal.

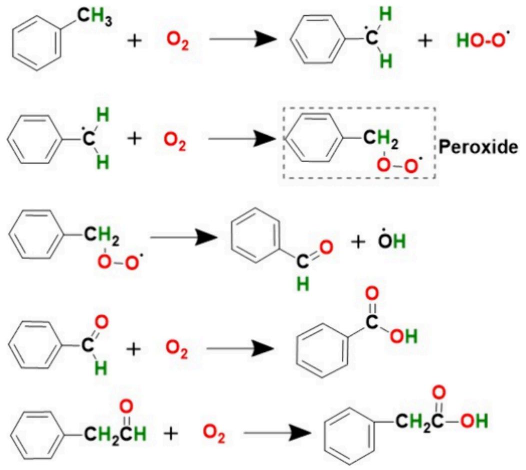

Figure 6. Oxidation reaction paths of functional groups with a side chain.

Coal pyrolysis commonly occurs under conditions of high temperature and oxygen deficiency. In addition to the decomposition of C=C bonds within the stable aromatic ring structure, it also leads to the cleavage of C−H and C−C bonds in the aliphatic side chain as well as a reduction in the content of C=O functional groups, which significantly influences the evolution of functional groups within the coal matrix during later stages of temperature increase. Throughout the heating process, a competitive relationship exists between these two reactions. During the initial phase of spontaneous combustion, when both the coal body temperature and heating rate are low while ambient oxygen content is relatively sufficient, changes in functional group content primarily result from oxidation reactions. At this stage (T1−T3), a large amount of active groups combine with oxygen, leading to a decreased oxygen concentration in the environment after this stage concludes; consequently, oxidation reaction processes slow down or even cease.

Figure 7 shows the content changes of aliphatic hydrocarbons (2950−2975 cm^{-1}, 2845−2870 cm^{-1}), hydroxyl groups (3200−3600 cm^{-1}) and ether bonds (1010−1275 cm-1) in the three coal samples during the temperature rise from room temperature 30 to 600 °C. Compared with raw coal, the content of aliphatic hydrocarbon side chains such as methyl and methylene is significantly reduced, indicating that flame retardants can effectively reduce the presence of active functional groups in the coal oxygen reaction, which may be due to the substitution of chloride ions on the aliphatic hydrocarbon side chains. Upon analysis of the coal samples treated with two types of flame retardants as depicted in the figure, it is evident that both chemical and bioretarded coal samples exhibit higher infrared absorption spectral intensity for aliphatic hydrocarbons compared to raw lignite coal. This suggests that even after undergoing reactions these impeded coal samples still contain intact aliphatic hydrocarbon side chains. It can be inferred that both flame retardants facilitate the solidification of C−H bonds in methyl and methylene functional groups within high temperature environments, effectively inhibiting their partic-

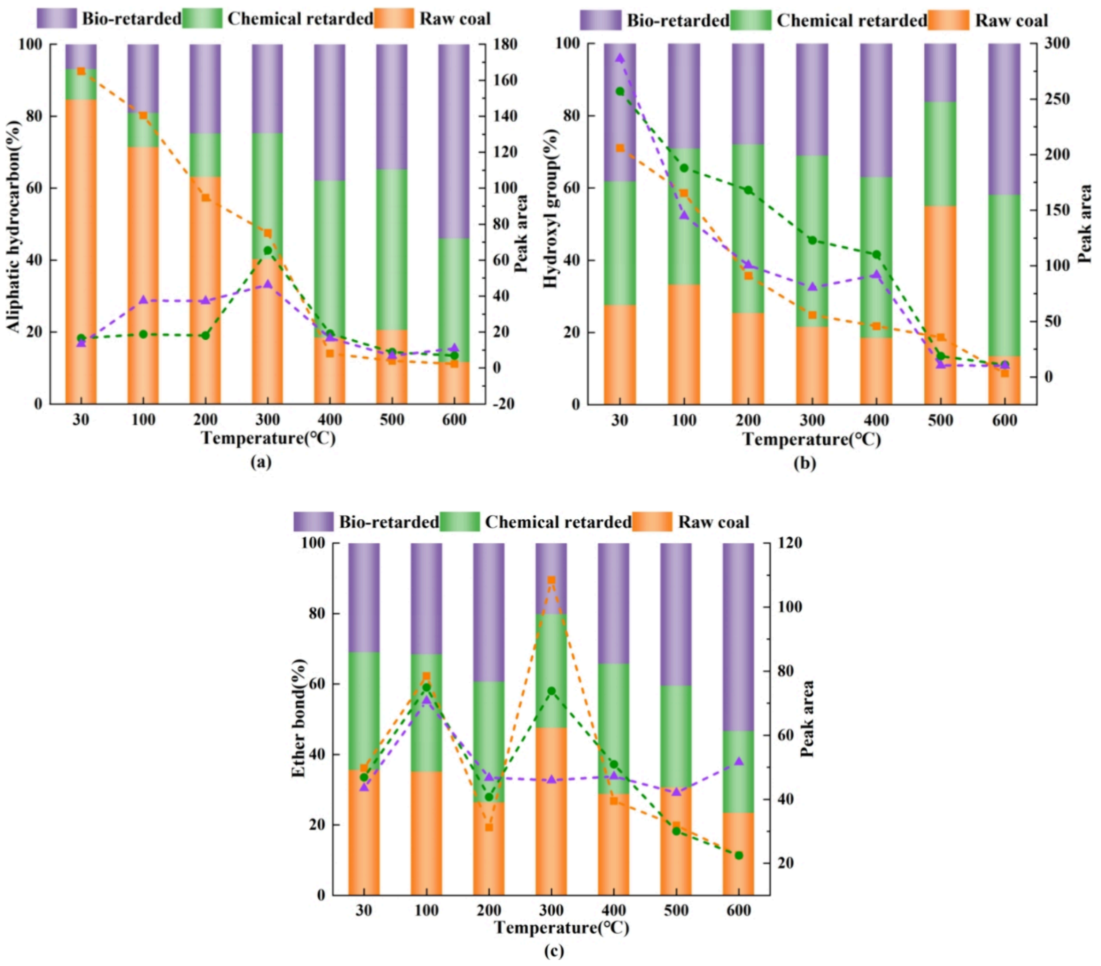

Figure 7. Changes of active group content in tested lignite samples at different temperatures.

ipation in subsequent oxidation reactions and delaying the involvement of aliphatic hydrocarbons in the oxidation process of coal to a certain extent. As a result, this influences the thermal properties and chemical stability of coal, as a natural polymer.

The continuous formation and consumption of O−H bonds occur in the whole heating process of coal. The change of hydroxyl content in coal at the early stage of low temperature oxidation is mainly caused by water evaporation caused by coal heating, which causes the chemical bond between hydroxyl and adjacent atoms to break, forming a double bond or other reaction products. With the further rise of temperature, groups such as carbonyl, carboxyl, methyl, and methylene oxidation and pyrolysis, resulting in the formation of associated hydroxyl groups, biobased flame retardants can effectively catalyze this reaction process, so at this stage, the content of free hydroxyl has a slight upward trend. At the same time, biobased flame retardants can promote the dehydrogenation of methyl and methylene groups on the side chain, allowing them to combine with oxygen to form stable ether bonds (R-O-R). This mechanism can effectively hinder the formation of carbon and oxygen intermediates to a certain extent, enhance the thermal stability of coal, and inhibit the spontaneous combustion of coal.

3.1.2. Analysis of Heat Release Characteristics. The TG-DTG-DSC curves of coal samples treated in different ways were obtained by analyzing the experimental data of the thermal gravimetric data of lignite, as shown in Figure 8.

As shown in Figure 8, the thermogravimetric (TG) curve demonstrates that the coal sample exhibits its highest weight loss rate when the temperature reaches 500−600 °C. This phenomenon can be attributed to an increased involvement of active groups in the coal-oxygen reaction in this temperature range, leading to the breakdown of the macromolecular structure of coal and subsequent gas generation. The characteristic temperature points and heat release data for both pre- and post-treatment of the coal sample are presented in Table 3.

It is evident from the observation table that, during the low temperature oxidation stage, the lignite treated with biobased flame retardant exhibits a delayed T3 temperature point at 292.94 °C, which is 71.62 °C later than that of raw coal. In the rapid oxidation stage, the characteristic temperature points of T4, T5, and T6 for biobased flame retardant lignite show an increase by 22.9, 61.31, and 89.09 °C, respectively. The inhibitory effect on the entire oxidation reaction process can be characterized by the mass loss rate. According to observations, compared to untreated coal samples, both types of flame retardants result in reduced quality loss rates in treated coal samples.

The application of chemical flame retardants results in a 12.42% reduction in the quality loss rate of coal samples, while the use of biobased flame retardants leads to a 15.05% decrease compared to untreated coal samples. This clearly demonstrates the significant inhibitory effect of biobased flame retardants on the oxidation and heat release processes in lignite.

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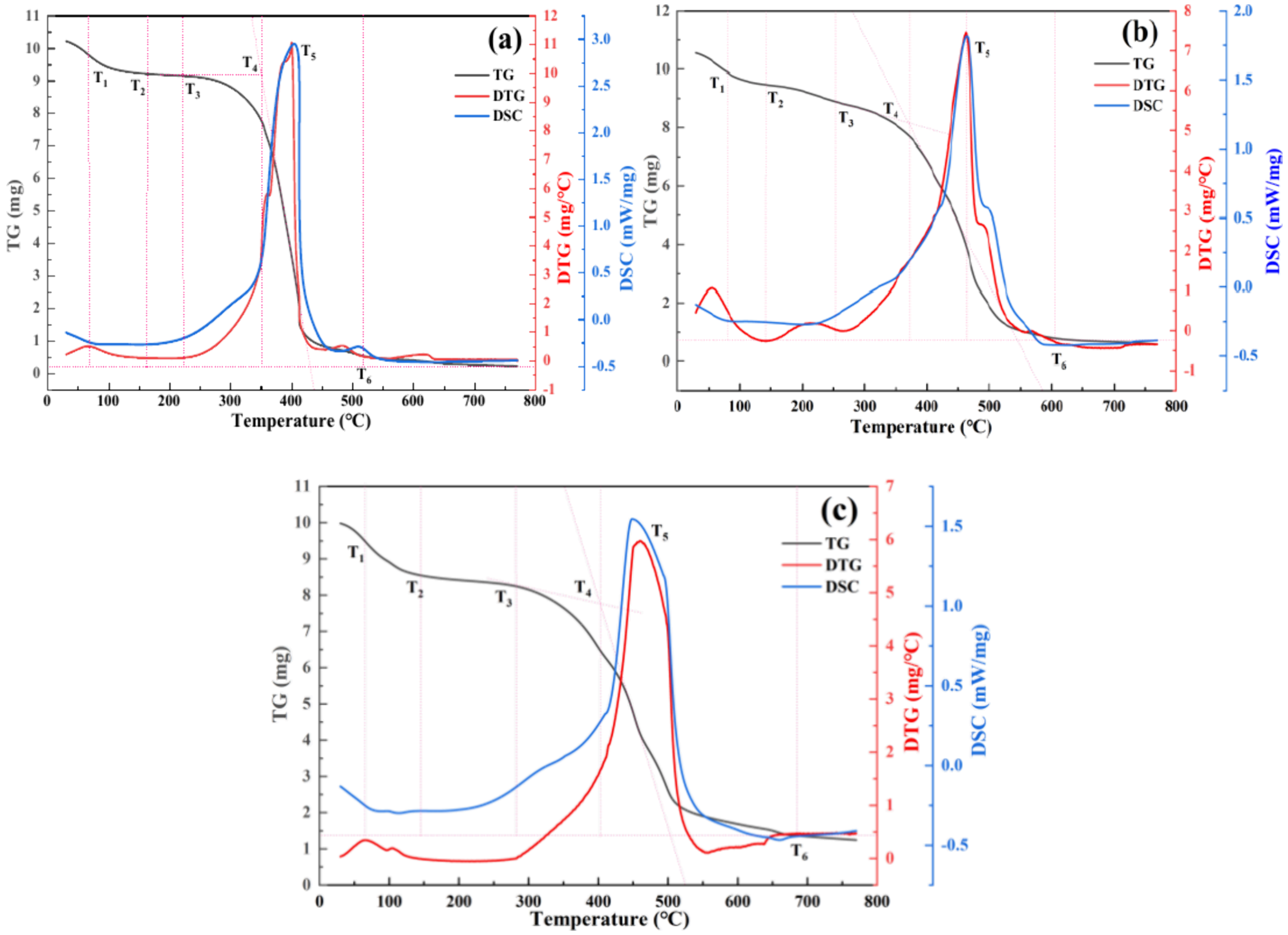

Figure 8. TG-DTG-DSC curve of tested lignite samples: (a) raw lignite, (b) chemical retarded lignite, and (c) bioretarded lignite.

Table 3. Characteristic Temperature Parameters of Tested Lignite Samples during the Coal Spontaneous Combustion Process

	Characteristic Parameters						
Coal Samples	T1	T2	T3	T4	T5	T6	Quality Loss Rate
raw coal	62.43	143.67	221.32	349.84	402.88	522.18	98.66%
bioretarded	80.08	140.25	292.94	372.74	464.19	611.27	83.61%
chemical retarded	66.82	145.56	282.21	403.48	449.57	605.45	86.24%
immersed sample	64.40	143.12	256.12	371.83	420.27	534.08	97.93%

According to the TG curve, the mass conversion percentage during the coal oxidation process can be represented as

$$\alpha = \frac{m_0 - m_t}{m_0 - m_f} \tag{13}$$

where m_0 is the initial mass value of the sample; m_t is the mass of the sample at a certain time; m_f is the mass of the sample remaining at the end of the reaction.

Figure 9 illustrates the correlation between the mass conversion percentage of lignite during thermal decomposition and combustion stages, following various treatments and temperature. The analysis reveals that the coal sample treated with biobased flame retardant necessitates a higher ambient temperature to achieve an equivalent conversion rate as the untreated coal sample. The thermal kinetics of coal aligns with its oxidation reaction since the biobased flame retardant generates calcium carbonate that adheres to the surface of the coal body, thereby reducing oxygen contact area and impeding the process of coal oxidation reaction.

3.1.3. Calculation of Oxidation Kinetics. The Coats-Redfern integral method was used to analyze the oxidation kinetics of different experimental coal samples during the oxidation process of thermal decomposition and combustion. The conversion rate ranges from 0.1 to 0.9. Table 4 shows the calculation data of the experimental process.[39,40] According to the calculation data in the table, $1000/T$ is taken as the horizontal coordinate and $\log[G(\alpha)/T^2]$ is taken as the vertical coordinate; the R of the fitting curve obtained is >20.97. Through the analysis of the fitting curve of the activation energy of the experimental coal sample, the activation energy E and the pre-exponential factor A can be calculated, as shown in Table 4.

As can be seen from Table 4, in the thermal decomposition stage, the activation energy of lignite after treatment with biobased flame retardants increased by 56.7%. In the combustion stage, it was increased by 153.4%. The activation

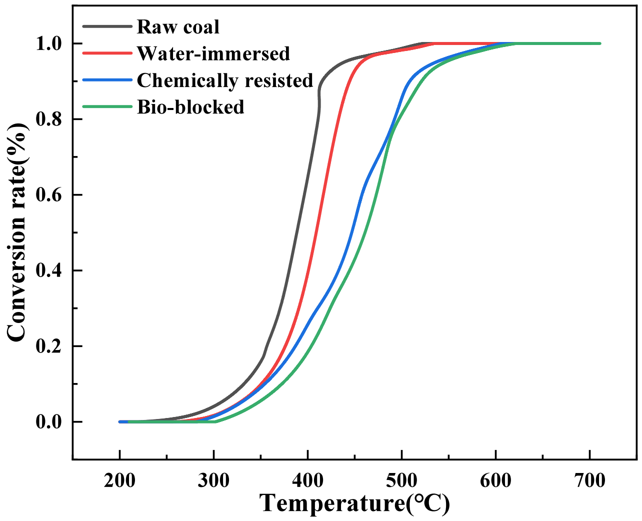

Figure 9. Conversion rates of tested lignite samples with the temperature.

energy of the bioretarded coal sample flame retardant consumes part of the oxygen-containing functional group in the coal molecule; the active group is reduced, and more heat is needed to activate, reducing the reactivity. The coal-oxygen reaction is more difficult.

3.2. Quantitative Test of Apparent Flame Retardancy.

3.2.1. Determination of Spontaneous Combustion Tendency. Based on the measured temperature and oxygen consumption at each crossing point of coal samples, the spontaneous combustion propensity of a coal sample can be calculated. The spontaneous combustion propensity of coal refers to its inherent oxidation capacity at room temperature, which reflects the degree of risk of spontaneous combustion during mining. This propensity primarily depends on factors such as the degree of coal metamorphism, brittleness, porosity, moisture content, composition of coal rock, and other internal factors. According to relevant national standards,[41] the classification index for assessing spontaneous combustion propensity should be based on the oxygen absorption per gram of dry coal at room temperature (30 °C) and atmospheric pressure (1.013 × 105 Pa). The temperature acquisition system is utilized for real-time monitoring of both coal and furnace temperatures. The intersection point temperature refers to the temperature at which the rise curve intersects.[42]

It can be observed from Figure 10 that, beyond 60 °C, diverse lignite samples exhibit distinct temperature rise rates. The coal samples treated with biobased flame retardation demonstrate a delay of 11.6 °C in comparison to the raw coal at the intersection point. Moreover, the cross-point temperature of the long flame coal sample is recorded as 152.3 °C, which indicates an 8.5 °C delay compared to the raw coal. It is worth noting that lower cross-point temperatures suggest a higher likelihood of spontaneous ignition for the respective coal sample.[43] Analyzing the relationship between intersection temperatures of different types of chemically and bioretarded coal samples reveals delayed temperatures when compared to untreated original samples. This is because the flame retardant product calcium carbonate functions by forming a protective layer on the surface of the coal seam, effectively isolating oxygen, and reducing the contact area between coal and oxygen. The reaction requires a higher temperature to activate the active groups. The calcium carbonate produced by biobased flame retardants exhibits greater stability compared to that generated by chemical flame retardants. Additionally, the presence of *Bacillus* in these biological agents contributes to their enhanced performance, as it acts as a viscous precipitated crystal nucleus for calcium carbonate, resulting in better cementation of coal seam particles. Consequently, the cross point temperature of coal samples treated with biobased flame retardants is higher than those treated with chemical counterparts.

According to eqs 9−12 obtained above, the oxygen consumption rate of experimental coal samples after different treatments is calculated. Figure 11 illustrates the oxygen consumption curves of lignite and long-flame coal. Prior to reaching 60 °C, there is no significant alteration in the oxygen content of both coal types. During this stage, physical adsorption and chemical adsorption are primarily responsible for oxygen consumption. As the temperature rises to 100 °C, there is a substantial increase in both the amount and rate of oxygen consumed by each coal sample. This can be attributed to an augmentation in active groups within the coal, which accelerates the reaction between coal and oxygen resulting in higher heat generation. These findings align with the observed changes in functional group content measured through in situ infrared experimentation conducted as part of this study. The oxygen consumption rate of the coal sample treated with biobased flame retardants exhibited a rapid increase at 110 °C, which was 10 °C lower than that of the untreated raw coal. At 150 °C, the oxygen consumption rate of the coal sample treated with biobased flame retardants was found to be lower compared to that of the coal sample treated with chemical flame retardants. This phenomenon can be attributed to the production of more stable calcite in the form of calcium carbonate by biobased flame retardants.

Moreover, microorganism-produced calcium carbonate acts as a superior cementing medium, exhibiting enhanced adhesion properties when compared to chemically produced calcium carbonate used in flame retardation. Consequently, this improved adhesive quality facilitates better interconnection between particles within the coal sample and results in an enhanced sealing effect. Based on the experimental results regarding the intersection temperature and oxidation characteristics of the two coal samples before and after flame retardancy, this article employs an identification method based on oxidation

Table 4. Oxidation Parameters of Tested Lignite Samples[a]

	Heat Decomposition Stage				Combustion Stage			
Coal Sample	E	A	$G(\alpha)$	R2	E	A	$G(\alpha)$	R2
raw coal	86.9	7.8	7	0.999	101.1	7.5	5	0.993
bioflame retardant	136.2	16.9	2	0.999	256.2	37.1	4	0.999
chemical retardant	133.1	16.9	5	0.964	235.4	34.7	5	0.998
immersed sample	102.8	10.4	7	0.998	185.9	22.1	5	0.993

[a] E is activation energy; A is prefactor, and $G(\alpha)$ is mechanism function.

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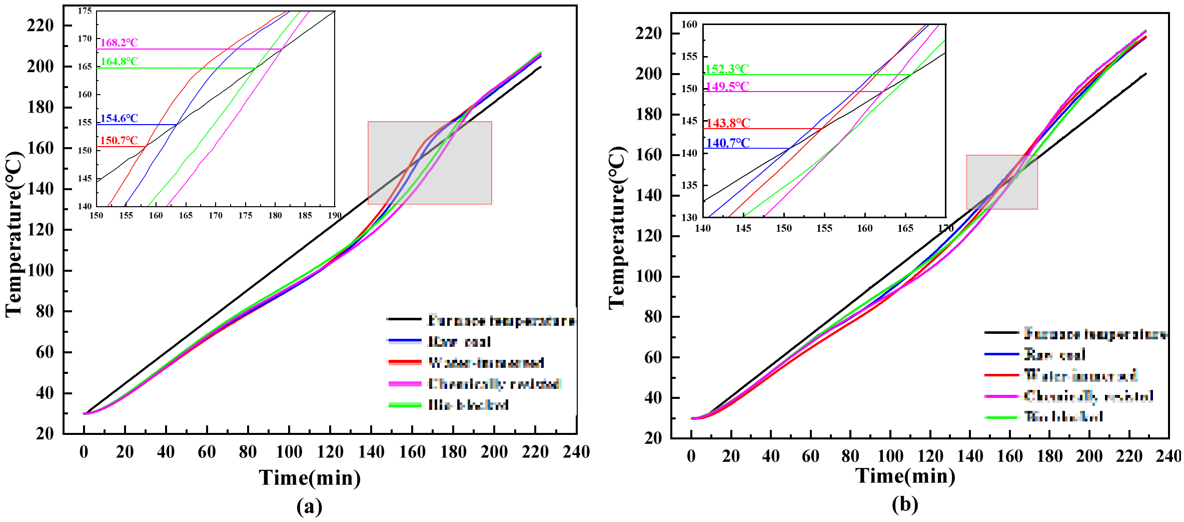

Figure 10. Comparison of intersection temperature of tested samples of (a) lignite and (b) long-flame coal.

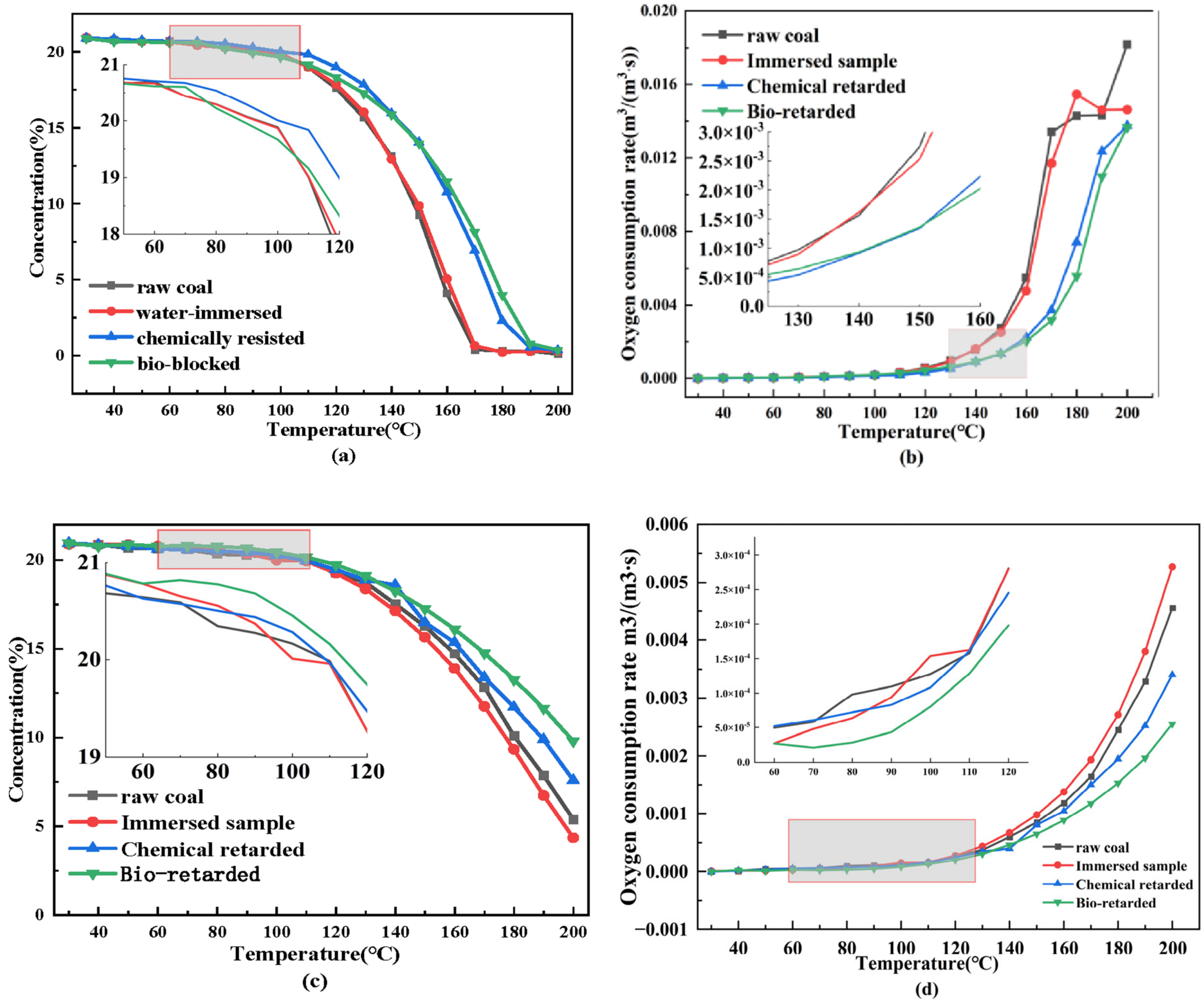

Figure 11. Oxygen consumption of tested coal samples at the low-temperature stage with different treatment methods: (a) oxygen concentration, lignite; (b) oxygen consumption rate, lignite; (c) oxygen concentration, long flame coal; (d) oxygen consumption rate, long flame coal.

Table 5. Identification of the Tendency Index of Spontaneous Combustion of Tested Coal Samples

Coal Sample					Decision Index		
		C_{O_2}	T_{cpt}	I	$I < 600$	$600 < I < 1200$	$I > 1200$
lignite	raw coal	20.43	154.6	631.7		√	
	bioretarded sample	20.61	168.2	812.8		√	
	chemical retardant	20.68	164.8	784.9		√	
	immersed sample	20.44	150.7	587.6	√		
long flame coal	raw coal	20.59	143.8	531.5	√		
	bioretarded sample	20.81	152.3	664.3		√	
	chemical retardant	20.57	149.5	594.4	√		
	immersed sample	20.65	140.7	506	√		

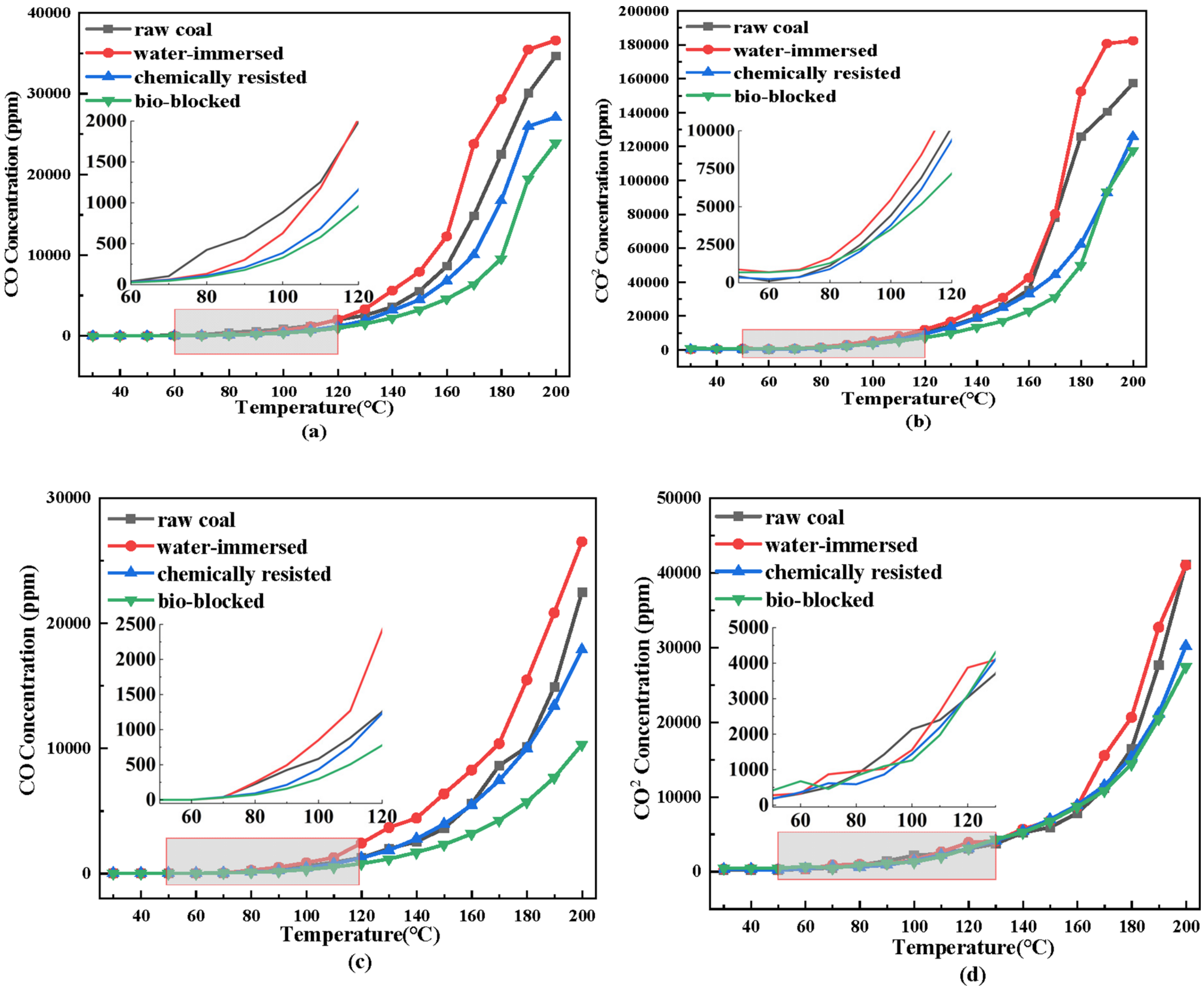

Figure 12. Changes in index gas production of tested coal samples at a low-temperature oxidation stage: (a) CO-lignite, (b) CO_2-lignite, (c) CO-long flame coal, and (d) CO_2-long flame coal.

kinetics to calculate a comprehensive evaluation index for assessing the tendency of coal spontaneous combustion. The formula utilized is as follows:

$$I_{C_{O_2}} = \left[\frac{(C_{O_2} - 15.5)}{15.5}\right] \times 100 \tag{14}$$

$$I_{T_{cpt}} = \left[\frac{(T_{cpt} - 140)}{140}\right] \times 100 \tag{15}$$

$$I = \phi(\psi_{C_{O_2}} I_{C_{O_2}} + \psi_{T_{cpt}} I_{T_{cpt}}) \tag{16}$$

where I is the judgment index of coal spontaneous combustion tendency, without dimension; I_{CO2} is the concentration index of oxygen in the outlet gas pipeline in the furnace when the coal temperature is 70 °C, without dimension; I_T is the temperature index of the cross point between the furnace temperature and the coal temperature, without dimension; C_{O2} is the oxygen concentration in the gas outlet pipe when the coal temperature reaches 70 °C, %; T_{cpt} is the temperature at the crossing point,

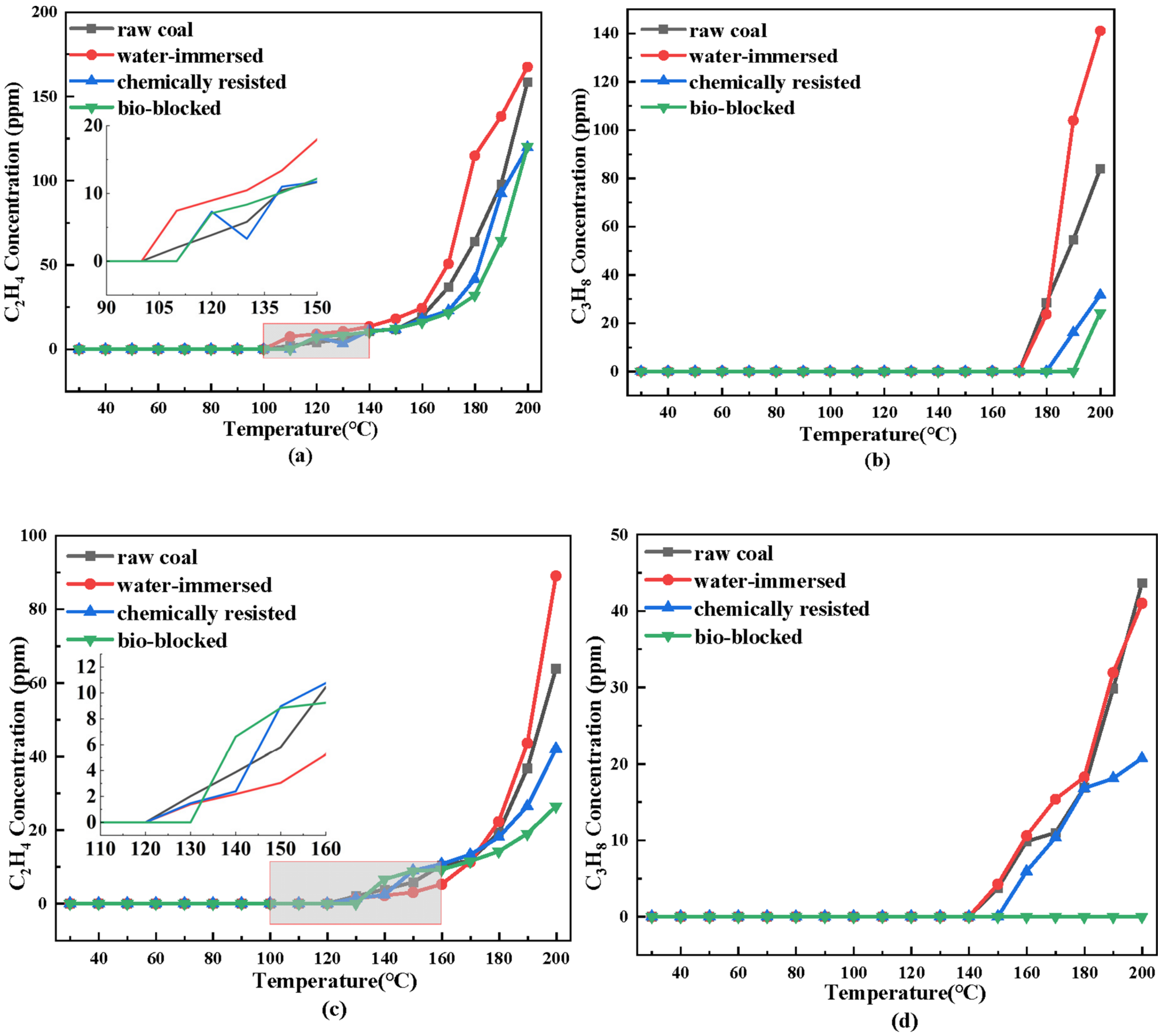

Figure 13. Changes in index gas production of tested coal samples at low-temperature oxidation stage: (a) C_2H_4-lignite, (b) C_3H_8-lignite, (c) C_2H_4-long flame coal, and (d) C_3H_8-long flame coal.

°C; 15.5 is the calculation factor of oxygen concentration at the outlet pipeline, %; 140 is the calculation factor of the temperature at the crossing point, °C; ψ_C is the weight of the low temperature oxidation stage, = 0.6; ψ_T is the weight of the rapid oxidation stage, = 0.4; ϕ is the magnification factor, = 40; 300 is the correction factor.

The results of the spontaneous combustion tendency calculation for lignite and long-flame coal are presented in Table 5. The natural inclination judgment index of lignite samples treated with chemical flame retardants and biobased flame retardants is 784.9 and 812.8, respectively, surpassing that of raw lignite coal. Notably, the utilization of biobased flame retardant on long-flame coal has altered its spontaneous combustion characteristics. According to the requirements stipulated in China's relevant coal mine fire prevention regulations, new mines that are susceptible to spontaneous combustion of coal seams should implement zonal ventilation or diagonal ventilation. Initially, central juxtaposed ventilation can only be employed in one mining area for production purposes, and this mining area must have at least one dedicated return air lane.[44] Furthermore, stricter management standards apply when the spontaneous combustion tendency index falls within the range of $1200 > I > 600$ for combustible briquettes. However, if biobased flame retardants are utilized to alter the classification of a coal mine's spontaneous combustion tendency, it can alleviate management difficulties to some extent while positively impacting the economic benefits associated with coal mining.

3.2.2. Flame Retardant Rate. According to the national coal industry standard, the flame retardant rate refers to the percentage of the CO volume concentration produced by raw coal sample and flame retardant coal sample when testing the flame retardant effect of flame retardants under specific laboratory conditions (MT/T 700−1997, 1997). According to the temperature rise experiment results set by the program, when the coal undergoes low temperature oxidation, a lot of oxygen and other harmful substances will be released; these substances include CO, CO_2, CH_4, C_2H_2, C_2H_4, C_2H_6, and C_3H_8.[45] In coal mining, you can estimate the temperature of the coal seam and the process of natural combustion by observing the concentration change of these iconic gases.

First, the impact of incorporating the biobased flame retardant into the tested coal samples on the generation characteristics of CO and CO_2 is illustrated. As can be seen from Figure 12, before the temperature reaches 60 °C, the generation of CO_2 and CO is not obvious, and the gas concentration produced is relatively low. After 80 °C, the concentrations of both gases begin to increase rapidly. For both lignite and long-flame coal, the CO and CO produced by the coal sample after chemical flame retardant and biobased flame retardant treatment are smaller than that of raw coal, and the temperature point at which the gas concentration begins to increase rapidly is also delayed compared to that of raw coal. At the same temperature, the gas concentration of the coal sample after biobased flame retardant treatment is smaller than that of the coal sample after chemical flame retardant treatment. This difference is more obvious in the concentration of the CO gas.

Additionally, C_2H_4 and C_3H_8 are used as representatives to analyze the effect of flame retardant treatment on the production of alkane gas in coal samples. As can be seen from Figure 13, the gas concentration generated by the coal samples of lignite and long-flame coal after flame retardant treatment is lower than that of raw coal during the heating process, and the gas concentration generated by the coal samples treated by the biobased flame retardant method is lower than that of the coal samples treated by the chemical flame retardant method at the same temperature. For lignite, the detection temperature of C_2H_4 and C_3H_8 of chemical flame retardant coal samples is 10 °C later than that of raw coal, and the detection temperature of C_3H_8 of biobased flame retardant coal samples reaches 190 °C, which is 20 °C higher than that of raw coal. When the temperature reaches 200 °C, the C_2H_4 concentration of both lignite and long-flame coal samples is lower than that of chemical flame retardant coal samples, and no C_3H_8 gas is produced in the whole low-temperature oxidation stage of biobased flame retardant long-flame coal.

According to the changes of CO gas production (Figure 12), the average flame retardant rate of lignite raw coal and coal samples treated by different flame retardant methods can be calculated, and the specific formula is as follows:

$$\varphi_r = \frac{Q1 - Q2}{Q1} \times 100\% \tag{17}$$

where φ_r is the flame retardancy rate of flame retardants to tested coal samples; Q1 is the release of CO from raw coal during the experiment; Q2 is the amount of CO released by the coal sample after flame retardant treatment during the experiment.

After calculation, it can be seen that the average flame retardant rates of lignite and long flame coal are shown in Table 6. It can be seen that, for lignite and long flame coal, the flame retardant rates of biobased flame retardants are 78.27% and 67.34%, respectively, showing good flame retardant properties.

Table 6. Flame Retardancy Rates of Tested Coal Samples

Coal Samples	Chemical Retardant	Bioretarded
lignite	74.18%	78.27%
long flame coal	59.42%	67.34%

4. CONCLUSION

The novel flame retardant material was successfully prepared through a meticulously designed experiment in this study. It was determined that the OD600 concentration in the biological culture solution is influenced by three crucial factors: culture temperature, culture time, and pH value of the culture environment. The recommended conditions for culturing include maintaining a temperature of 25 °C for 48 h with an environmental pH level of 7.5. Furthermore, to obtain high-quality calcium carbonate in MICP products, it is suggested to use a cementing/bacterial fluid concentration ratio of 1:1.1 and a urea/calcium source ratio of 1:1.5.

This study comprehensively tested the entire heat release process, from low temperature oxidation to high temperature combustion, of raw coal and biologically retarded coal samples. The study also discussed the reaction mechanism of a new retardant agent that inhibits spontaneous combustion in coal. The results indicate that the addition of biobased flame retardants can consume methyl and methylene in coal, effectively inhibiting its pyrolysis during heating. This promotes the formation of stable ether bonds in coal and slows its oxidation process. Thermogravimetric analysis revealed that, compared to raw coal, the temperature delay for bioflame retardant lignite at stage T3 was 60.89 and 71.62 °C, respectively, while the characteristic temperature point (T4−T6) for coal samples treated with bioflame retardant increased by 22.9, 61.31, and 89.09 °C, respectively, during rapid oxidation stages. The characteristic temperature points of chemically inhibited lignite were increased by 53.64, 46.69, and 83.27 °C, respectively. The results of oxidation kinetics analysis indicated that the activation energy of lignite treated with biobased flame retardants increased by 56.7% in the thermal decomposition stage compared to raw coal. In the combustion phase, the increase was even more significant, reaching 153.4%. This phenomenon can be attributed to the consumption of oxygen-containing functional groups in the coal molecules by the biobased flame retardants, resulting in a reduction in active groups and an increased requirement for additional heat to activate the reaction.

The inhibitory performances of biobased flame retardants on lignite and long flame coal, two typical low-rank coals, were quantitatively analyzed by calculating the changes in components during spontaneous combustion. The evaluation index for the tendency of spontaneous combustion significantly increased to 812.8 for lignite treated with biobased flame retardants compared to the raw coal's value of 631.7. For long flame coal, the application of biobased flame retardants transformed its spontaneous combustion tendency from easy to moderate. Based on the CO concentration curve as an indicator gas, the flame retardancy achieved by biobased flame retardants for lignite and long flame coal reached 78.27% and 67.34%, respectively, demonstrating commendable fire-retardant properties.

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Author Contributions

Conceptualization, Y.W., X.C., and S.W.; formal analyses, Y.W. and R.L.; writing—original draft, R.L. and S.W.; writing—review and editing, Y.W., X.Z., and D.L. All authors have read and agreed to the published version of the manuscript.

Notes

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