



Experimental studies of spontaneous combustion and anaerobic cooling of coal



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HIGHLIGHTS

- A 15-t experimental furnace of coal spontaneous combustion was constructed.
- Characteristics of coal spontaneous combustion and anaerobic cooling were studied.
- Temperature variations of coal spontaneous combustion and anaerobic cooling were obtained.
- Quantitative calculation method of oxygen consumption rate and heat intensity was created.
- Corresponding relationship between gas ratios and coal temperature was established.

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ABSTRACT

To study the spontaneous combustion and anaerobic cooling of coal, a 15-t experimental furnace was constructed, and large-scale spontaneous combustion tests were conducted. The coal sample came from No. 3 seam in Dongtan mine in Yanzhou coalfield, Shandong province, China. The variations in temperature, gas ratios, oxygen consumption rate, and heat intensity were studied. The experimental results show that for coal spontaneous combustion below 70 °C, the temperature increases slowly with time and the oxygen consumption rate and heat intensity increase slowly with temperature and accelerate when the temperature exceeds 100 °C. In anaerobic cooling, the cooling rate decreases with the decrease in temperature. When the coal temperature goes from 452.7 to 250.0 °C, the cooling rate decreases rapidly, whereas below 250.0 °C, the cooling rate decreases slowly. The variation trends of the CO₂/CO, CH₄/C₂H₆, C₂H₄/C₂H₆, and C₃H₈/C₂H₆ gas ratios in the spontaneous combustion and anaerobic cooling of coal in a certain range of temperature can be used to predict the temperature of coal.

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1. Introduction

Coal fire is one of the key hazards in coal mines, among which that caused by the spontaneous combustion of coal is significant. In the major coal-producing countries, such as China, USA, India, and Indonesia, a large amount of coal reserves are destroyed every year by coal fires. Some coal seams smolder for decades. During the low-temperature oxidation and combustion of coal, certain greenhouse gases such as CO₂, CH₄, and other toxic gases (CO, SO₂, etc.) are produced, causing environmental pollution. Coal waste is also prone to spontaneous combustion with a negative impact on the environment [1–5]. Spontaneous combustion of coal is a complex physicochemical process, including physicochemical adsorption

and chemical reactions with heat release. When heat is accumulated, the temperature increases, thus accelerating the reactions with oxygen and leading to coal spontaneous combustion when the temperature reaches ignition temperature.

The spontaneous combustion of coal is influenced by many factors, mainly including oxygen partial pressure, particle size, moisture content, and temperature [6]. Temperature has a pivotal influence on the spontaneous combustion of coal. The increase in coal temperature accelerates the formation of CO₂, CO and other oxidation products, and the oxygen consumption rate almost doubles for a temperature rise of 10.0 °C [7]. Oxygen is the key factor for the spontaneous combustion of coal. The reactive functional groups in coal react with oxygen, and the heat released from the reactions is accumulated, thus increasing the temperature. As reported previously, the oxygen consumption rate enhances with the increase of oxygen partial pressure when the pressure is less than 1 MPa. With a constant oxygen partial pressure, the lower

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Nomenclature

C_{O_2}	oxygen concentration (%)	$V_{O_2}^0(T)_{ref}$	reference oxygen consumption rate of coal in fresh air ($\text{mol s}^{-1} \text{mm}^{-3}$)
$C_{O_2}^0$	standard oxygen concentration (%)	r	radial coordinates
$C_{O_2}^i$	oxygen concentration of a certain point (%)	S	furnace sectional area (mm^2)
$C_{O_2}^1, C_{O_2}^2$	oxygen concentrations of two points (%)	T	temperature ($^\circ\text{C}$)
c_c	coal specific heat capacity ($\text{J g}^{-1} \text{ }^\circ\text{C}^{-1}$)	Z	vertical coordinates
c_g	air specific heat capacity ($\text{J g}^{-1} \text{ }^\circ\text{C}^{-1}$)	z_i	distance of a certain point to the entrance (mm)
c_e	coal equivalent specific heat capacity ($\text{J g}^{-1} \text{ }^\circ\text{C}^{-1}$)	z_1, z_2	heights of two points (mm)
d_{ref}	reference particle size (mm)	a	volume of mole vapor under standard conditions ($2.24 \times 10^7 \text{ mm}^3 \text{ mol}^{-1}$)
d_{50}	average particle size (mm)	Greek symbols	
n	porosity	τ	reaction time (s)
Q	experimental supply air volume ($\text{mm}^3 \text{ s}^{-1}$)	$\phi(d_{50})$	influence function of particle size
\bar{Q}	air leakage intensity ($\text{mm}^3 \text{ mm}^{-2} \text{ s}^{-1}$)	λ_c	coal thermal conductivity ($\text{J mm}^{-1} \text{ s}^{-1} \text{ }^\circ\text{C}^{-1}$)
$q(T)$	heat intensity ($\text{J s}^{-1} \text{ mm}^{-3}$)	λ_g	air thermal conductivity ($\text{J mm}^{-1} \text{ s}^{-1} \text{ }^\circ\text{C}^{-1}$)
$q_0(T)$	heat intensity when the oxygen concentration is standard ($\text{J s}^{-1} \text{ mm}^{-3}$)	λ_e	coal equivalent thermal conductivity ($\text{J mm}^{-1} \text{ s}^{-1} \text{ }^\circ\text{C}^{-1}$)
$V_{O_2}(T)$	oxygen consumption rate ($\text{mol s}^{-1} \text{ mm}^{-3}$)	ρ_c	coal density (g mm^{-3})
$V_{O_2}^0(T)$	oxygen consumption rate of coal in fresh air ($\text{mol s}^{-1} \text{ mm}^{-3}$)	ρ_g	air density (g mm^{-3})
		ρ_e	coal equivalent density (g mm^{-3})

the total oxygen pressure, the higher the oxygen consumption rate [8]. In the early stage of the low-temperature oxidation of coal, the reaction activation energy decreases with the increase in oxygen concentration, whereas in the final stage of oxidation, when the oxygen concentration is below a certain value, the reaction activation energy increases, inhibiting oxidation [9]. Therefore, fires can be prevented and extinguished by reducing the oxygen concentration. Particle size significantly affects the area where the coal contacts with oxygen.

When the particle size is smaller, the area is larger, which facilitates coal oxidation [10,11]. The effect of water on the spontaneous combustion of coal is complex. On one hand, coal absorbs water, releasing moist heat, condensation heat, and hydrolysis heat, promoting coal oxidation. On the other hand, water affects the diffusion rate of oxygen, and water evaporation absorbs heat at the same time, inhibiting coal oxidation [12]. Other materials present in coal and additives also affect the spontaneous combustion of coal. For example, sulfur generally exists in coal in the form of FeS. When FeS is oxidized, heat is released, and an acidic environment is created. In addition, porosity of coal increases due to inflation [13]. All these affect the spontaneous combustion of coal obviously. Some additives promote or inhibit spontaneous combustion. For example, KAc, Ca(Ac)₂, and NaAc will promote spontaneous combustion, while NaCl and CaCl₂ have a negative influence on spontaneous combustion [14]. Some inhibiting additives retard the spontaneous combustion of coal obviously by changing the surface structure of coal in chemical methods, and the inhibition is even several times higher than that by pure water, such as urea [15]. Therefore, researchers have studied the application technology of inhibitors on the fire prevention and control [16,17].

The mechanism of coal oxidation and modeling of the dynamics of spontaneous combustion are vital fields. In the low-temperature oxidation of coal, oxygenated complexes are generated, and two parallel reaction sequences contribute to the emission of carbon oxides during oxidation [18]. The formation of activated oxygenated complexes and their subsequent thermal decomposition play an important role in the low-temperature oxidation of coal with changes in the five element occurrences in coal matrix, and H-containing groups are more easily react with molecular O₂ to form hydroxide intermediates [19]. The studies of diffusion mechanism indicate that the diffusivity of oxygen is associated with

porosity and particle size in the range from 10⁻⁸ to 10⁻⁶ m² s⁻¹. The oxygen diffusivities of large and fine particles are mainly governed by continuum diffusion and Knudsen diffusion/kinetics diffusion, respectively [20].

The coal molecular structure is one of the key intrinsic factors for the spontaneous combustion of coal. The functional groups in coal molecular structure react with oxygen and release heat. Different molecular structures of coal have different quantities and types of reactive functional groups, and at the same time, the solid oxides formed by the low-temperature oxidation of coal also affect the spontaneous combustion of coal [21]. Infrared spectroscopy is commonly used to investigate the molecular structures and oxidation products of coal. Studies have shown that hydroxyl is one of the key functional groups during the spontaneous combustion of coal: the higher the degree of coal metamorphism, the lower the retractable strength [22]. The absorbance of aliphatic hydrogen decreases with temperature rise. The absorbance of oxygen-containing functional groups increases gradually with temperature rise and then decreases because of the decomposition of oxygen-containing functional groups or reactions with other functional groups or oxygen when the temperature reaches above a certain value [23]. Because of the problems in technical and data processing of infrared spectra, some researchers have proposed quantum chemistry analysis based on Beer–Lambert law and used the Gaussian to fit data, achieving sound results [24].

Thermogravimetry is generally used with other methods to study the spontaneous combustion of coal and investigate the kinetics parameters [22,25]. Large-scale experiments on the spontaneous combustion of coal are efficient methods for studying spontaneous combustion processes. By simulating the spontaneous combustion of coal, characteristic parameters, such as temperature, oxygen consumption, gas variation, heat intensity, and the limiting parameters, can be determined and the combustion zone can be located more accurately. Furthermore, reasonable index gases can be selected to predict the spontaneous combustion of coal [26]. Index gases should be sensitive to temperature or demonstrate a special variation trend at a certain temperature. Moreover, the gases should be easily detected. For example, ethylene is generated when the temperature of coal reaches a certain point. However, because of the low concentration and the dilution with ventilation air, it is difficult to detect ethylene until

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