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Thermodynamic characteristics of coal reaction under low oxygen concentration conditions

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1. Introduction

ABSTRACT

In order to further understand the characteristics of coal reaction under low oxygen concentration atmosphere, this study tested the thermodynamic characteristics during reaction processes under low oxygen concentrations. The kinetic factors were also analyzed based on testing results. The results show that there are some characteristic temperatures during coal spontaneous combustion and the influences of oxygen concentration on these temperatures are different. The influences of oxygen concentration on the mechanism function and kinetic parameters depend on the coal ranks and the reaction stages. They are different at different reaction stages for the same coal, and at the same reaction stage for different coals. This phenomenon is essentially resulted from the different influences of oxygen concentration on the micro reaction sequences during coal spontaneous combustion. The study will be helpful for further understanding of the actual development process of coal spontaneous combustion.

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The spontaneous combustion of coal often occurs during the processes of coal mining, storage or applications. Almost all the coal mining countries are facing the danger of coal spontaneous combustion [1-4]. In addition to burning of coal resource, different types of harmful gases are generated during the process of coal spontaneous combustion, which will seriously threaten people's safety and environment. Under some special conditions, coal spontaneous combustion may result in serious gas or dust explosions.

The spontaneous combustion of coal is resulted from the accumulation of heat released from coal reaction. So the thermodynamic characteristics in different stages of coal spontaneous combustion are important for further understanding of this disaster. Many investigators have done a lot of studies on the heat production and dynamic parameters during coal spontaneous combustion. Most of these previous investigations focused on the coal reaction under oxygenic or dry-air conditions. In contrast, few literatures have been published on the topic of thermodynamic characteristics under low oxygen concentration conditions. By the method of TG, some researchers investigated the thermogravimetric changes, exothermic properties and thermodynamic parameters during coal spontaneous combustion process under the conditions of dry-air, high concentration oxygen or pure oxygen [5-14]. Based on the TG testing results, some investigators analyzed the effects of different oxygen concentration on the reaction degree, ignition time and characteristic temperatures of coal [15–18]. However, most of these conclusions are qualitative and cannot provide enough data for further understanding coal reaction under low oxygen concentration conditions. By the method of DSC, some researchers investigated the heat release characteristics of coal reaction under the conditions of dry-air or pure oxygen atmosphere, and proposed some determining methods for the propensity of coal to spontaneous combustion [19–22]. By the method of DTA, Pis et al. [23] tested the thermal characteristics of coal samples oxidized for different times under air atmosphere at 200 °C and also proposed determining methods for the propensity of coal to spontaneous combustion. Haykiri-Acma et al. [24] applied DTA and DTG techniques investigated combustion characteristics of bituminous coal samples, found that volatile matter and fixed carbon contents of the investigated samples on a dry-ash free basis were seen to have a strong effect on the cumulative mass loss values. Marinov et al. [25] used TGA/DTA to study the changes in the combustion behavior of microbial treated

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coals. In recent years, some new thermal analysis instruments began to be used in the field of coal spontaneous combustion. Using C80 Micro-calorimeter, Wang et al. [26,27] and Qi et al. [28] researched the exothermic characteristics of coal oxidation at temperatures ranging from room temperature to 300 °C, found that the oxygen consumption was proportional to the amount of heat release. Using Pulse Calorimeter, Li et al. [29,30] analyzed the thermodynamic properties of the adsorption heat, condensation heat and evaporation heat of coal during coal oxidation process at low temperatures. In addition, some investigators studied the thermal characteristics of coal oxidation through the analysis based on chemical theory [31–34]. Xu [31,32] inferred the distribution range of exothermic intensity of coal oxidation at low temperatures according to oxygen consumption, heat production rate and bond energy change, and also analyzed the influence of oxygen concentration on the exothermic intensity of coal oxidation [33]. Cheng et al. [34] deduced the thermal effect during coal oxidation at low temperatures under dry-air atmosphere through the analysis based on chemical theory.

In summary, the previous investigations on thermal effects of coal reaction were usually carried out under the atmospheres of pure oxygen, dry-air or other sufficient oxygen supply conditions, while the investigation under low oxygen atmosphere is few. And these existing few investigations taken under low oxygen concentration conditions cannot provide enough data for further understanding coal reaction under low oxygen concentration conditions. Actually, most of the coal spontaneous combustion zones, such as goaf, loose coal bulk, sealed fire zone, coal pile, coalfield fire zone, etc., usually occur under the conditions of low oxygen concentration below 15%. Therefore, the existing research results of coal reaction obtained under the experimental conditions of sufficient oxygen supply cannot reflect the actual process of coal spontaneous combustion, and cannot provide an efficient guidance for the prevention of this hazard.

This study analyzed the thermodynamic characteristics during the reaction processes of three different ranks of coal samples under different oxygen atmospheres by the thermogravimetric analyzing method. The results will be helpful to further understand of the actual development process of coal spontaneous combustion occurring during the processes of coal mining, storage or applications.

2. Experimental

2.1. Coal samples

Three ranks of raw coal were collected from different coalfields in China, i.e. Changcun jet coal, Jiaxiang gas coal and Zhaogezhuang fat coal. They have different propensity to spontaneous combustion. Based on the statistics, their shortest times to spontaneous combustion are 7 days, 25 days and 182 days. The coal samples were processed to remove the surface layers and the material was then crushed in an oxygen-free glove box. This resulted in the coal particles ranging between 0.18 mm and 0.38 mm in size which were sieved and used as the experimental coal samples. The coal particles were kept under an inert atmosphere before they were tested. Considering on the effects of storage on coal self-heating [35,36], the coal samples should be tested no longer than one week. The proximate analyses of the samples are listed in Table 1.

2.2. Testing facility

The testing system includes SDT-Q600 synchronous thermal analyzer, MF-4 gas mixture, pure nitrogen bottle, pure oxygen bottle and computer (see Fig. 1). The synchronous thermal analyzer can continuously test TG curves during coal reaction process. The pure nitrogen bottle and pure oxygen bottle are connected with gas mixture through gas tube, which form a high precision gas mixture system. This gas mixture system can provide mixed N_2 – O_2 gas with different oxygen concentrations by setting the gas pressure, gas flow rate and mixing ratio of pure nitrogen and pure oxygen. So the coal reaction processes under the atmospheres at different oxygen concentrations can be simulated accurately.

During the testing process, the coal sample will be put into the coal reaction vessel. Then we turn on the instruments and set their running parameters, such as initial temperature, final temperature and temperature rising rate. At the same time, let the mixed gas with different oxygen concentrations to flow into the coal reaction vessel. The thermal analysis data during coal reaction under different oxygen atmosphere will be collected on-line until reaching the final temperature.

2.3. Testing procedure

First, 8 mg of coal particles were packed into the coal reaction vessel. Then the programmed temperature program was set to run at a programmed heating rate of 5 °C/min while mixture gas of oxygen and nitrogen with different oxygen concentration was permitted to flow through the coal reaction vessel at a rate of 100 mL/min. The temperature was set rising from room temperature to 1000 °C. The oxygen concentration used in this study include 5%, 7%, 10%, 13%, 16%, 19% and 21%. During the temperature rising process, the instrument continuously monitored the thermodynamic data and show with changing curves.

Table 1

Proximate analyses (air-dry basis) of coal samples.

Coal samples	M (wt%)	A (wt%)	V (wt%)	FC (wt%)	Q _{net} (MJ/kg)	H (wt%)	S _t (wt%)
Jet coal	9.86	20.65	30.01	34.08	21.81	4.49	0.37
Gas coal	2.82	7.85	34.82	54.51	31.89	3.21	0.21
Fat coal	1.20	17.48	26.71	55.10	28.48	4.59	1.16

Notes: M – moisture content; A – ash content; V – volatile matter; FC – fixed carbon; Q_{net} – net calorific value; H – hydrogen content; S_t – total sulfur content.

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