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Risk evaluation of coal spontaneous combustion on the basis of auto-ignition temperature

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ABSTRACT

The spontaneous combustion of coal, if not eradicated immediately, may lead to coal ignition and even a full-blown fire. A new method, DSC Inflection Point (DSCIP), was proposed to determine the coal auto-ignition temperature (CAIT). Heat fluxes and kinetic parameters before and after CAIT were comparatively investigated through TG/DSC analysis and mathematical model construction. Meanwhile, the impacts of temperature rise rate and oxygen concentration on CAIT were studied and two indexes representing the hazard and destructiveness of coal spontaneous combustion, respectively, were proposed. The results demonstrated that the heat flux curve of coal spontaneous combustion can be well fitted using Gaussian mixture model. Compared to the oxidation stage, the released heat during the combustion stage was greatly increased. Furthermore, the activation energy became larger and the reaction order decreased to zero when the temperature exceeded CAIT. The study also found that CAIT was enhanced with the increase of temperature rise rates or the decrease of oxygen concentrations. Changes of heat flux, free radicals, and the activation energy proved the rationality and feasibility of the DSCIP method in determining CAIT. Additionally, under the same environmental conditions, lignite had the largest hazard of coal spontaneous combustion and the anthracite had the biggest destructiveness. Both the hazard and the destructiveness of coal spontaneous combustion became stronger as oxygen concentrations increased.

1. Introduction

The spontaneous combustion tremendously affects the normal extraction of coal. If not immediately attended to, the spontaneous combustion may engulf a large area and spread further. This will not only endanger miners and mining operations, but also result in possible mine closures and fatalities [1–3]. CAIT can provide guidelines for preventing, controlling and monitoring the spontaneous combustion. CAIT is defined as the minimum temperature at which a coal ignites spontaneously under the conditions without external ignition source [4]. However, the determination of CAIT is still controversial. Oxidant addition method was employed to measure CAIT in China's National Standard of 'Determination of Ignition of Coal' [5]. In this method, Sodium Nitrite and Benzidine with an appropriate mixing ratio are added into the coal sample prior to each test and a deflagration should be observed during the test. CAIT is determined as the corresponding temperature when the deflagration occurs. Basically, two phenomena indicate the occurrence of deflagration: a sudden expansion of the gas volume in the reaction system or a sudden rise of the coal temperature. However, the deflagration might not be clearly measured for coals containing high ash content. In addition, different testing results will be obtained for various devices, operating methods or $NaNO_2$ additions. As a result, this testing method is not extensively used in practice.

In recent years, thermogravimetric analysis and thermal analysis technologies were widely used to determine CAIT due to its time saving convenience. Muthuraman et al. [6] defined CAIT based on the temperature at which the DTG had its peak value and the corresponding slope to the intersection with respect to the TG profile. Li et al. [7] used the intersection of two tangents along a TGA curve to determine CAIT. Lu and Chen [4] compared the two-tangent method with deviation method and found that the ignition temperature increased with increasing heating rates. Luo et al. [8] found that ignition temperatures decreased with the increasing oxygen concentrations through TG tests. According to thermal analysis, Kim and Jeon [9] used the temperature at which the d*T*/d*t* of the coal surface reached to the minimum to represent the ignition point. Zhang et al. [10] determined the ignition temperature by the point where the heat release rate of carbon oxidation is equal to the endothermic heat flow. Li et al. [11] used the temperature of the net exothermic heat flow to indicate the onset of coal oxidation and the temperature corresponding to the DDSC peak to

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represent the fastest coal oxidation. By summarizing the methods from the literatures, the following methods were popularly used.

(1) Thermal Equilibrium Method (TEM): This method was proposed by International Confederation for Thermal Analysis (ICTA) who recommended that CAIT could be determined by the transition point from endothermal to exothermal processes during coal oxidation [12].
(2) Mass Loss Percentage Method (MLPM): In this method, CAIT is established by the point where the mass loss percentage of coal sample reaches 10% on thermogravimetric analysis (TG) curve [13].
(3) Oxidation-Pyrolysis Division Point (OPDP): CAIT is determined by the temperature at which the mass loss curve of the coal combustion is separated from the curve of coal pyrolysis [14].
(4) Peak Mass Point (PMP): During the combustion process, the mass of coal sample initially decreases and gradually increases to a peak, then drops down rapidly. The corresponding temperature where the coal mass increases to the peak is considered as CAIT [15].
(5) TG Tangent Method (TGTM): The corresponding temperature of the intersection between the base line and the tangent line of the maximum mass loss rate point on TG curve is considered as CAIT [16,17].

The temperature determined by TEM is actually the transition from moisture evaporation/gas desorption stage to oxidation stage. At this temperature, only a few combustible species are released so as not to support the ignition of coals. PMP and OPDP are roughly the onset points of coal pyrolysis. In fact, ignition takes place after the concentration of combustible gases in the air reaches to a threshold. So, ignition is not able to occur at PMP or OPDP due to the insufficient combustible gases. Meanwhile, different coal types have various properties and thus the 10% mass-loss point cannot represent the ignition characteristics for all of the coals. Additionally, MLPM and TGTM are actually the results based on mathematical treatments. The auto-ignition temperature is essentially a transition point from one state to another and therefore the point used to determine CAIT should be a critical point or a jump on a curve, which, obviously, cannot be satisfied by MLPM or TGTM.

Through the analysis above, the methods introduced from literatures might not be able to represent CAIT, which necessitates an accurate determining method in order to help prevent and control the spontaneous combustion of coal.

2. Determination of CAIT by DSC inflection point (DSCIP)

The combustion process of coal generally contains two stages or types: the volatile component combustion stage and the char combustion stage [18]. The release of volatile components starts from the end of moisture evaporation/gas desorption stage (roughly equivalent to the temperature determined by TEM). Subsequently, the coal surface starts to absorb and react with oxygen, simultaneously releasing heat and oxidation species including CO, CO_2, H_2O and so forth. As the heat accumulates, other gaseous substances such as CH_4, C_2H_4 etc. will be liberated. But at this stage, coal temperature and the concentrations of gaseous substances are not enough to trigger the ignition. After the temperature reaches OPDP, a large number of volatile components start releasing. When the temperature and the concentration of the volatile components both exceed certain values, ignition might occur. Once the coal is ignited, the released heat and the combustion products will significantly increase. As a result, CAIT should be characterized by:1) higher than OPDP; and 2) a critical point exhibiting disparate combustion properties before and after it.

The phenomena observed during the spontaneous combustion of coal primarily contain mass loss, gases liberation and heat release, among which the former two are actually the external reflections of coal oxidation, while released heat is the essential factor leading to spontaneous combustion according to Semenov Combustion Theory and Frank-Kamenetskii Combustion Theory [19,20]. Mathematically, an inflection point is usually regarded as a transition point, at which the trends or properties change. In this case, the inflection point on DSC curve is proposed to represent CAIT (marked as DSCIP method). Accordingly, the whole spontaneous combustion process can be divided into four stages based on the DSC curve: dehydration/desorption stage (from the beginning to TEM), oxidation stage (from TEM to CAIT), combustion stage (from CAIT to the peak on DSC curve) and decaying stage (from the peak on DSC curve until the end). Oxidation stage is characterized by slow chemical reactions and can take place at low temperatures as long as the energy of the system exceeds the activation energy of a chemical reaction [21]. However, combustion can only occur at strict conditions. According to Fire Triangle Theory, the coal can be ignited only when the temperature, the oxygen concentration and the concentration of the released combustible gaseous products during coal oxidation all exceed certain values. Since CAIT is the watershed of oxidation stage and combustion stage, the features of these two stages were studied using TG/DSC and Electron Spin Resonance (ESR) technique to investigate the combustion parameters before and after CAIT.

3. Experimental setup and procedures

3.1. Sample selection and preparation

Three coal samples were collected from different coal mines and denoted by Coal A, Coal B and Coal C, respectively. Coal A is lignite, Coal B bituminous coal and Coal C anthracite. The properties of the three selected coals were tested by 5E-MAG6700 Automatic Industrial Analyzer and 5E-CHN2200 Elemental Analyzer. The results were displayed in Table 1. Each coal sample, as received, was crushed, pulverized and sieved to less than 0.098 mm. The fine coal particles were then kept in dry and sealed glass bottles for tests.

3.2. Thermogravimetric experiments

The schematic of the experimental system used in thermogravimetric tests was illustrated in Fig. 1. The system primarily contains four parts: the gas supply system, gases mixer, TG/DSC analyzer, and a computer. The gas supply system, including a nitrogen bottle and an oxygen bottle, is used to supply N_2/O_2 mixture with various ratios. The gases mixer can guarantee the uniformity of the mixed gases. Coal samples are oxidized in the TG/DSC analyzer. With the proceeding of the coal spontaneous combustion, the mass loss and heat release rate

Table 1
Proximate analysis and ultimate analysis of the coal samples.

Coal Samples	Proximate Analysis (%)				Ultimate Analysis (%)				
	Moisture	Volatile Matter	Ash	Fixed Carbon	C	H	O	N	S
Coal A	9.17	39.03	13.32	38.48	54.85	2.97	13.68	2.65	0.58
Coal B	4.44	27.90	11.28	56.38	71.47	3.58	10.35	1.48	0.25
Coal C	1.89	7.69	25.37	65.05	82.13	3.67	2.05	1.44	0.27