Paper No. 29

ISMS 2016

Influence of Fundamental Internal Parameters to Low-Temperature Critical Temperature in Coal Self-Ignition Process

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ABSTRACT
The low-temperature oxidation process of coal self-ignition can be divided into two stages: the slow oxidation stage and the fast oxidation stage. To study the effect of internal factors on low-temperature critical temperature in the coal self-ignition process, eight fundamental internal parameters (volatile, ash, fixed carbon, C, N, S, H, and O) and three characterization parameters of low-temperature critical temperature in the coal self-ignition process (T_{JR}, $T_{CO,}$ and T_{O2}) were determined through theoretical analysis. Afterwards the self-heating characteristics in pure oxygen atmosphere in the 40 to 180° range and gas releasing atmosphere in the 30 to 180° range for seven coal samples were tested. Based on this, the values of fundamental internal parameters and characterization parameters for seven coal samples were obtained. The relationship between fundamental internal parameters and that of characterization parameters were compared. On grey relational analysis, five important influence parameters are filtered to make the mathematical formula fit with the characterization parameters. The results show that characterization parameters reflect critical temperature from different aspects including temperature rising, CO releasing, and oxygen consumption. These vary for the same coal sample, but are with the same tropism and confirm each other. Critical temperatures are negatively correlated with the contents of volatiles, H and O, have no determined relationship with the content of S, and have small correlation with other parameters.
Keywords: coal self-ignition; critical temperature; fundamental internal parameter; temperature-programmed experiment; heat oxidation experiment; grey correlation; numerical fitting.

1. INSTRUCTIONS

The occurrence of coal self-ignition is the result of the interaction between external and internal factors, and among them, the internal factors are prerequisites (Liu and Zhou, 2012; Adamus et al., 2011). Low-temperature critical temperature is the intermediate link between the slow oxidation stage and fast oxidation stage during the coal self-ignition process. It is of practical significance for doing coal mine fire prevention and control that we grasp the critical temperature of coal self-ignition and make the temperature of coal below the critical temperature. In the past, we primarily focused on monitoring the coal temperature of danger zones through direct temperature measurement or gas collection and testing in terms of coal mine fire prevention (Xie et al., 2011). However, there is little research and application of nonlinear characteristics of spontaneous fires. Currently, some research is being done on the determination of critical temperature of coal self-ignition mainly through fixed stages division, qualitative analysis, and the numerical fitting method (Sahay et al., 2007; Zhong et al., 2010; TAN et al., 2013; Beamish and Darren et al., 2005; Ren et al., 1999; ZHU et al., 2014). These methods mainly analyze the critical temperature of coal self-ignition in one aspect. Researching the critical temperature form different aspects and contacting the nature and conditions of coal are necessary in order to increase the practical applications and reference significance.

The occurrence and development of the coal self-ignition process can be manifested by different forms, such as changes in temperature, gas concentration, and so on. In this paper, a relationship between coal temperature and gas concentration is established by experiments, the critical temperature of coal self-ignition is determined by different parameters, and the influence of some fundamental parameters of coal, such as industrial components and element composition, on critical temperature are studied.

2. THEORETICAL ANALYSIS

Coal self-ignition is a non-linear evolution process, which is not carried out in accordance with a

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certain rate. Research shows that there is critical temperature during this process where the oxidation of coal will accelerate beyond this temperature. However, the critical temperature of coal is limited within a fixed range with no clear distinction between the different kinds of coal. In recent years, scholars have studied in depth the differences of coal self-ignition process for different types of coal, and come to realize the importance of distinguishing the differences in critical temperature. According to the previous studies, the influence of internal and external factors on the occurrence and development of coal self-ignition is very complicated, and the influence of external factors is difficult to be experimentally tested. Therefore, it is of guiding significance to actual work to study the influence of internal factors on critical temperate of coal self-ignition. Therefore, this paper mainly studies the impact of fundamental internal parameters on low-temperature critical temperature of coal self-ignition.

2.1 Fundamental internal factors

The study of coal self-ignition mechanisms shows that the type and quantity of functional groups plays a key role in the coal self-ignition process (Wang et al., 2012; Satoru et al., 2000; Olayinka, 1992). However, it is difficult to explain the difference between functional groups of different types of coal quantitatively, and not operable in engineering practice. Various functional groups in coal are constituted by the respective elements, industrial ingredient is a side reaction of coal structure and composition of functional groups, element composition and industrial components are the basic parameters of coal, and can easily be tested. Therefore, industrial components (volatile, ash, and fixed carbon) and element composition (C, H, O, N, and S) are selected as influence factors, and the influence of these factors on low-temperature critical temperature in the coal self-ignition process is analyzed. It should be noted that the moisture content of coal is not selected as one influence factor in this paper, because coal samples were dried prior to the experiment.

2.2 Characterization parameters of low-temperature critical temperature

The coal self-ignition process shows varying characteristics, especially at the critical temperature. Changes of coal temperature and gas concentration have a large effect on the coal self-ignition process, so this paper uses the following parameters as characterization parameters for low-temperature critical temperature.

1) Mutation temperature of heating rate in the adiabatic oxidation process (T_{JR}), which reflects the variation characteristics of coal heating rate under adiabatic conditions.

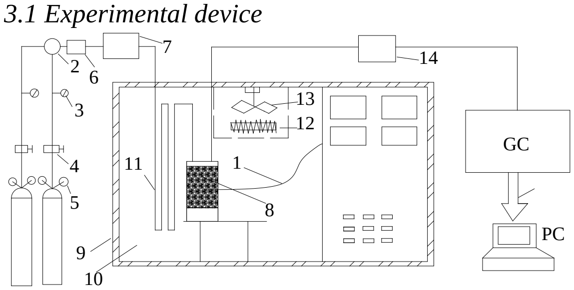
2) Mutation temperature of CO concentration in the programmed temperature rising experiment (T_{CO}). CO is a sort of important indicator gases during coal self-ignition process, the concentration of CO can be used to characterize the reaction rate, so the mutation temperature of CO concentration can be used to characterize the critical temperature.

3) Mutation temperature of oxygen consumption rate in the programmed temperature rising experiment (T_{O2}). Oxygen consumption rate is a direct exhibit of the reaction rate, so oxygen consumption rate can be used to characterize the critical temperature.

To study the influence of fundamental internal parameters to critical temperature of coal self-ignition, this paper will study the relationship between the characterization parameters and the internal parameters (T_{JR}, T_{CO} and T_{O2}).

3. EXPERIMENTAL PART

3.1 Experimental device



1-Temperature sensor 2-Three-way valve 3-Pressure gauge 4-Regulator valve 5-Pressure reducing valve 6-Dust collector 7-Mixing chamber for inlet gas 8-Reaction vesse l 9-Thermal insulation layer 10- Temperature control oven 11-Preheat copper pipe12-Heating pipe 13-Fan 14-Mixing chambe for outlet gas

Figure 1: Structure schematic diagram of the device.

This device consists of four parts: thegas distribution part, reaction vessel, temperature control oven, and control part. Figure 1 shows the structures schematic diagram. The design of this device used the adiabatic oven of the University of Queensland as a reference (Beamish et al., 2000). The temperature control oven and its door are double skinned, and there is glasswool in it for insulation. A 2.3 kW heating pipe is used for heating, and there is a fan to force gas flow to ensure temperature uniformity inside the oven. There is a 16 m length of copper pipe in the oven to ensure the temperature of gas is the same as the oven before entering the reaction vessel. Taking the vacuum flask as the adiabatic reaction vessel can effectively reduce conduction and radiation heat transfer. The temperature programmed reaction vessel is made of steel to ensure good thermal conductivity, and type K thermocouples are used for monitoring temperature. The gas used in the experiment is provided by high-pressure gas cylinders, and the gas is control by mass flow meter after flowing through the pressure valve. The control part is a PC, and the device is controlled by King View software. The flow of gas and temperature of the oven can be regulated and the date can be recorded by operating this software.

3.2 Experimental procedure

1) Preparation of coal samples

The coal samples used in this study were obtained from coal mines of different regions in China. All the coal samples selected for testing were obtained directly from bore holes, and then put into a plastic bag. After this, the coal samples were put into a low-temperature closed container and transported to the laboratory, then placed into a refrigerator until the start of the experiment. Before testing, the coal samples were divided into two parts (nearly the same): one part was returned to the refrigerator after rewrapping. The other part was sieved by jaw crusher and closed grinding machine to select expected particle size for immediate testing.

2) Experimental procedure

(1) Temperature programmed experiment

A 200 g coal sample was crushed into particles with mixed diameters of 1.25-1.6 mm, 1.6-2 mm, 2-3.5 mm, 3.5-5 mm and 5-7 mm, each accounting for 20%. The coal samples were put into the reaction vessel, dried for 9 h in nitrogen environment (120 ml/min) at 105°. After cooling down, nitrogen-oxygen mixed gas (the concentration of oxygen is 21%) was transported into the programmable isothermal oven with a constant flow rate of 60 ml/min. Emission gases were collected once every 15°, and the oxygen concentration was analyzed with a gas chromatograph; the initial temperature and termination temperature were 30° and 180°. In the heating process, temperature rise inside the oven was 15° with a rate of 1°/min, and the temperature kept constant for 10 min, and then increased 15° with a rate of 1°/min again. Similar procedures were carried on until 180°.

(2) Adiabatic oxidation experiment

The granularity of coal samples are <212 μm. All coal samples were dried at 105° for 15 hours under the protection of nitrogen to ensure that the coal samples were dried completely and not oxidized. Then the temperature was cooled down to 40° and allowed to equilibrate. The drying coal sample was moved into the adiabatic reaction vessel quickly and stabilized at 40° under nitrogen atmosphere, then switched to “track temperature mode”. The nitrogen input was cut off and open oxygen valve with a constant flow rate of 60 ml/min. The change in coal temperature was recorded by the control part for later analysis. When the coal temperature reached 180°, heating and the oxygen flow were automatically stopped. The coal sample was removed from the adiabatic reaction vessel after the sample and oven were cooled down to normal atmospheric temperature. The vessel was cleaned and the adiabatic device was checked.

3.3 Experimental results

Based on the experimental method above, the concentration-temperature curve of O2 and CO of temperature programmed experiment and temperature-time curve of adiabatic oxidation experiment for seven coal samples can be obtained, which are shown in Figures 2-4.

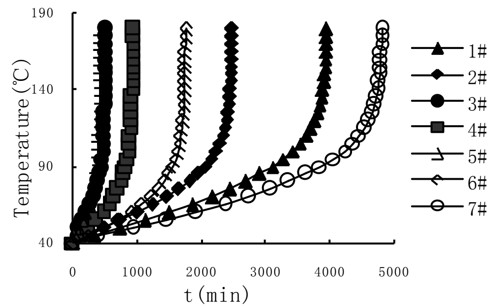


Figure 2: The coal temperature-time curve in isolated heat oxidation experiments.

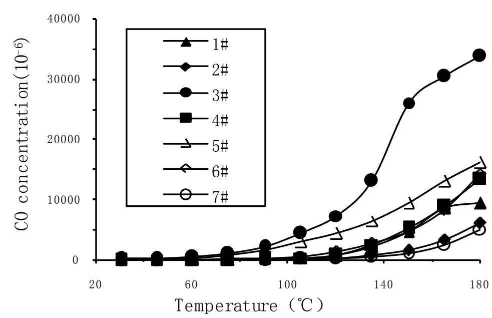


Figure 3: The change of CO concentration with temperature improving in temperature-programed experiments.

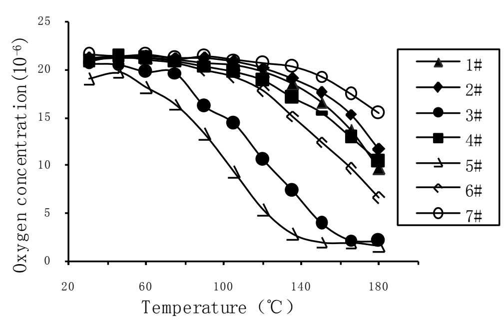


Figure 4: The change of O_2 concentration with temperature improving in temperature-programed experiments.

3.4 Oxygen consumption rate analysis

The height of the reaction vessel is 0.25 m (coal sample height is 0.221 m, L), Cross-sectional area (S) is 0.007088 m^2. The temperature of the reaction vessel is considered to be changed evenly for low heating rate and light coal sample weight. It is assumed that the gas used is ideal gas. Depending on the coal oxidation reaction equation (Srinivasan et al., 1996; Copard et al., 2004), oxygen consumption rate equation of physical coal per unit length at a certain position (x) is given by Formula (1):

$$-\upsilon_g dC^x_{o_2} = (1-n)R^x_{o_2}(T)Sdx \tag{1}$$

where n is porosity, %; S is cross-sectional area of the reaction vessel, m^2; $R^x_{o_2}(T)$ is oxygen consumption rate at position x, mol/(m^3·s); υ_g is gas flow rate, m^3/s; $C^x_{o_2}$ is oxygen concentration at position x, mol/m^3; $dC^x_{o_2}$ is variation of the oxygen concentration, mol/m^3.

According to chemical kinetics and equilibrium theory (Kudynska and Buckmaster, 1996; Wang et al., 2003; Baris et al., 2012), the average oxygen consumption rate can be expressed as Formula (2).

$$R_{O_2}(T) = R^x_{O_2}T\frac{Ci}{C^x_{O_2}} \tag{2}$$

where $R_{O_2}(T)$ is the average oxygen consumption rate, mol/(m^3·s); C_i is oxygen concentration of inlet gas, mol/m^3. Substituting equation (2) into equation (1), equation (3) reads:

$$R_{O_2}(T) = \frac{\upsilon_g Ci}{(1-n)SL}\ln\left(\frac{Ci}{C^L_{O_2}}\right) \tag{3}$$

Depending on equation (3), the values of RO2 for seven coal samples are given in Figure 5.

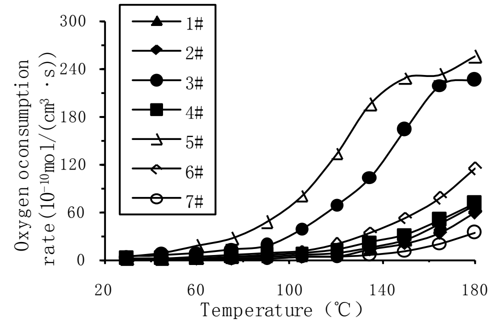


Figure 5: The change of oxygen consumption rate with temperature improving in temperature-programed experiments.

4. PARAMETER CALCULATION AND ANALYSIS

4.1 Internal parameters

Element contents (N, S, H, O, and C) and industrial components (ash, volatile, and fixed carbon) of seven coal samples are tested by vario MACRO CHNS (with Oxygen kit) Elemental analyzer and GF-A6 Automatic Industrial Analyzer. The results are shown in Table 1.

Table 1: The result of industrial analysis experiments and element analysis experiments.

	Volatile property wt% ar	Ash wt% ar	Fixed carbon wt% ar	N wt% daf	S wt% daf	H wt% daf	O wt% daf	C wt% daf
1#	21.42	19.86	57.52	1.185	1.179	3.761	6.332	69.99
2#	25.63	22.72	50.32	1.206	1.01	3.818	8.396	65.29
3#	33.18	8.84	52.57	1.309	2.495	4.814	15.11	69.16
4#	39.15	9.55	46.95	1.239	0.582	4.111	15.92	70.12
5#	30.23	11.34	52.68	1.148	0.929	4.234	14.59	84.98
6#	30.88	3.7	61.45	1.281	0.852	3.834	9.614	75.72
7#	12.95	8.07	77.72	1.332	0.634	3.731	3.321	69.81

3.2 Characterization parameters of critical temperature

According to Arrhenius equation (Bews et al., 2001; Ronald et al., 1989; Anna et al., 2011), reaction rate can be express as:

$$k = A\exp(-\frac{E}{RT}) \tag{4}$$

where k is Oxidation reaction rate of coal; E is apparent activation energy, KJ·mol^{-1}; A is pre-exponential factor, s^{-1}; R is universal gas constant, 0.008314 KJ/(K•mol); T is thermodynamic temperature, K.

Further analysis of the formula (4):

$$\ln k = \ln A - \frac{E}{RT} \tag{5}$$

Replace the Oxidation reaction rate k with adiabatic heating rate, CO concentration and oxygen consumption rate (denoted as k_1, k_2, and k_3), the relationship between lnk and (-1/T) can be obtained. Using linear analysis, the critical temperature can be obtained by analyzing the changes of slope, which is shown in Figures 6-8.

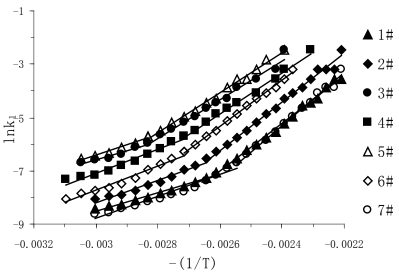


Figure 6: The relationship between $\ln k_1$ and (-1/T).

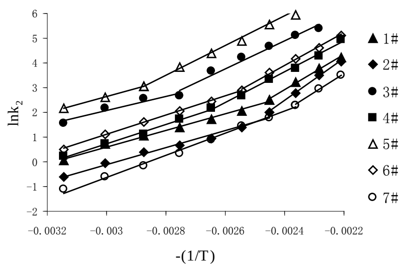


Figure 7: The relationship between $\ln k_2$ and (-1/T).

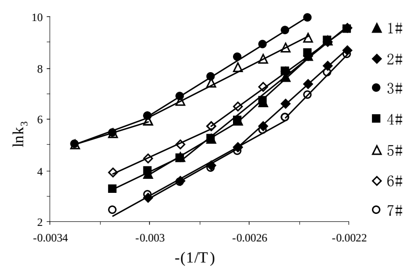


Figure 8: The relationship between $\ln k_3$ and (-1/T).

According to the analysis results above, critical temperatures for seven coal samples are shown in Table 2.

Table 2: Characterization parameters of coal spontaneous combustion critical temperature.

Item	T_{JR} (℃)	T_{CO} (℃)	T_{O2} (℃)
1#	110	105	135
2#	105	105	135
3#	80	60	90
4#	95	75	105
5#	80	60	75
6#	95	90	120
7#	120	135	150

5. INFLUENCE OF INTERNAL PARAMETERS TO CRITICAL TEMPERATURE

5.1 Relationship between internal parameters

The relationship between the eight parameters is shown in Figure 9. Based on this, it can be seen that the relationship between the various parameters is complicated; however, it still can be clearly observed that the trend of O and volatile property is similar, and the trend of fixed carbon is contrary to them.

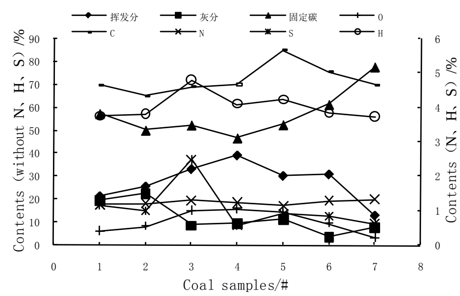


Figure 9: Fundamental internal parameters and the relationship.

5.2 Relationship between characterization parameters

The relationship between the three characterization parameters is shown in Figure 10.

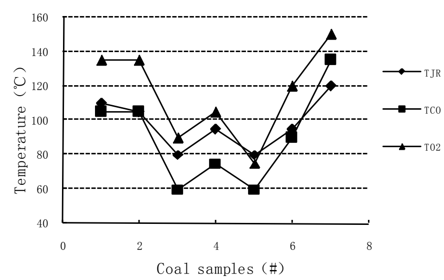


Figure 10: Characterization parameters of coal spontaneous combustion critical temperature.

According to Figure 10, the changing trends of the three parameters for different coal samples are basically the same. In accordance with the ascending parameter values, coal numeral order is 5#, 3#, 4#, 6#, 2#, 1#, and 7#. The 3 parameters for the same coal sample are different, for example, T_{JR}, T_{CO} and T_{O2} for 1# coal sample are respectively 110, 105 and 135, which shows that the changing regulations for the three characteristics are the same, but there is little differences in temperature. Overall, values of the three parameters for a certain coal sample are very close and are valid parameters.

5.3 Gray relational grade analysis

The relationship between internal parameters and characterization parameters of critical temperature is complicated, and the influence is hard to obtain by direct observation. Based on this feature, this paper introduces the gray relational grade analysis method to analysis the relevance of eight internal parameters and three characterization parameters. The calculation is according to the literature (LIU et al., 2004), and the calculation methods and formulas are not given in this paper. Based on data from Table 1 and Table 2, the calculation result is given in Table 3.

Table 3: Gray relational grade between characterization parameters and fundamental internal parameter.

	T_{JR}	T_{CO}	T_{O2}	Mean value
Volatile property	0.68	0.69	0.65	0.67
Ash	0.55	0.70	0.64	0.63
Fixed carbon	0.67	0.56	0.54	0.58
N	0.56	0.62	0.56	0.58
S	0.70	0.58	0.54	0.61
H	0.59	0.62	0.59	0.60
O	0.65	0.66	0.62	0.64
C	0.54	0.54	0.52	0.53

According to Table 3, the three highest relevance parameters are contents of volatile property, ash, and O, the lowest ones are contents of fixed carbon, C, and N. In decreasing order: volatile property, O, ash, S, H, fixed carbon, C, and N, which reflects the influence level of parameters.

5.4 Numerical fitting analysis

To further analyze the influence of internal parameters to characterization parameters, this paper makes a mathematical formula fitting to those two sorts of parameters. The fitting expression is labelled formula (10). As in the analysis above, the contents of fixed carbon, C, and N are the lowest relevance parameters. They do not have an obvious influence on the characterization parameters, and therefore we only consider the three characterization parameters and five internal parameters (volatile property, O, ash, S, and H).

$$y = p_0 (x_1)^{p_1} (x_2)^{p_2} (x_3)^{p_3} (x_4)^{p_4} (x_5)^{p_5} \quad (10)$$

where y is 3 characterization parameters T_{JR}、T_{CO} and T_{O2}, x_1-x_5 are 5 internal parameters, volatile property, O, ash, S and H, p_0-p_5 are fit coefficients.

Using the date in Tables 1 and 2, the fitting result is shown in Table 4.

Table 4: Numerical fitting results.

	p_0	p_1	p_2	p_3	p_4	p_5
T_{JR}	430.13	-0.14	-0.004	0.17	-0.50	-0.14
T_{CO}	376.39	-0.23	0.05	-0.07	-0.17	-0.27
T_{O2}	2317.83	-0.11	-0.04	0.28	-1.54	-0.19

According to Table 4, p_1, p_4 and p_5 for the numerical fitting of three characterization parameters are negative, which shows that the critical temperature and contents of volatile parameter, O, and H are negatively correlated; the absolute values of p_2 are very small, which shows that critical temperature has very little correlation with the content of ash; p_2 and p_3 are positive or negative, which shows that the influence of contents of ash and S is not certain.

6. CONCLUSIONS

To study the effect of internal factors on low-temperature critical temperature in coal self-ignition process, eight fundamental internal parameters (volatile, ash, fixed carbon, C, N, S, H, and O) and three characterization parameters of low-temperature critical temperature in the coal self-ignition process (T_{JR},T_{CO} and T_{O2}) are selected, and the values for each are determined via experiment and calculation. Then, the relationship between fundamental internal parameters and that of characterization parameters are compared. Through grey relational analysis, five important influence parameters are filtered to make a mathematical formula fitting with the characterization parameters. The conclusions are as follows:

(1) Based on the theoretical analysis of the occurrence and development of coal self-ignition, T_{JR}, T_{CO}, and T_{O2} are selected as characterization parameters of critical temperature, which characterize critical conditions for three aspects: heat rising, CO generation, and O_2 consumption.

(2) Depending on the Arrhenius equation, the values of characterization parameters for seven coal samples were obtained by the segmented fitting method, which shows that the changing regulations for the three characteristics are the same, but there is little difference on temperature; values for the three parameters for a certain coal sample are very close, and are valid parameters.

(3) By analyzing the relevance of characterization parameters and fundamental internal parameters, it is found that the critical temperature and contents of volatile parameter, O, and H are negatively correlated; critical temperature has very little correlation to the content of ash; the influence of contents of ash and S are not certain, and the influence of contents of fixed carbon, C, and N to critical temperature is the least significant.

(4) The accuracy of parameters and the number of coal samples need to be improved to get more accurate results in future research.

7. ACKNOWLEDGEMENT

The authors gratefully acknowledge the financial support provided by National Nature Science Foundation of China (U1261214) and Development Fund Project of China Coal Research Institute (2014JC06).

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