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An approach for evaluation of grading forecasting index of coal spontaneous combustion by temperature-programmed analysis

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Abstract

Focused on the prediction and forecast index of coal spontaneous combustion, the temperature-programmed experiments were carried out to identify the releasing rule of the gaseous compounds with the consideration of coal particle sizes. Based on the method of growth rate analysis, critical and dry cracking temperature temperatures were determined, dividing the coal low-temperature oxidation process into three stages. The critical temperature for the spontaneous combustion of the coal is in the range of 60°C~70°C, with the dry cracking temperature ranging from 100°C to 110°C. It was

found that the particle sizes had obvious effects on the release of gaseous compounds during the coal heating test. To reduce the influence of coal particle size on the forecasting of coal spontaneous combustion, the combination methods of coefficient of variation and the weighted grey relational analysis were proposed for the selection and evaluation of coal spontaneous combustion indexes. For the tested coal sample, CO_2/CO was selected as the first index, $\text{O}_2/(\text{CO}+\text{CO}_2)$ and CO_2/O_2 as the second index. C_2H_4 and C_2H_6 were selected as the index gases to confirm that the coal oxidation reached the dry cracking temperature. This research could provide a reference for the determination of the grading forecasting index of coal spontaneous combustion.

Keywords: Coal spontaneous combustion; Particle size; Grading forecasting index; Coefficient of variation; Weighted grey relational analysis

1. Introduction

Coal spontaneous combustion is one of the major disasters that threaten the safe production of coal mines (Taraba and Michalec 2011; Kong et al. 2017). It not only causes wastage of resources (Shao et al. 2015; Wang et al. 2018a; Li et al. 2016), but also leads to gas and coal dust explosions, which threaten the health of mineworkers seriously. In addition, hazardous gas, as well as greenhouse gases, are generated (Wang et al. 2016; Xu et al. 2018; Honscha et al. 2021), thus, causing significant damage to the atmosphere and ecological environment (Wang et al. 2018b; Zhang et al. 2016; Song and Kuenzer 2014). Accurate prediction and forecasting are important to control the hazard of coal spontaneous combustion (Onifade and Genc 2020; Qu 2018). In this respect, the determination of the quantitative index to predict and forecast the coal spontaneous combustion in a complex environment becomes a research hotspot (Yuan and Smith 2011; Baris et al. 2012).

During the coal spontaneous combustion, various gas products are released, and their concentrations are positively correlated with coal temperature (Ma et al. 2019). These gases are usually used to predict and forecast coal spontaneous combustion, which is very important for mine safety (Deng et al. 2014a, 2015). Guo et al. (2019a) assessed the qualitative and quantitative analysis of the CO and C₂H₄ formation rates, as well as various gas ratios to forecast coal spontaneous combustion. Deng et al. (2014b) proposed the growth rate analysis method to identify the characteristic temperature of spontaneous combustion. Guo et al. (2019b) found that the olefin index gas can predict coal spontaneous combustion more accurately than alkane index gas in low oxygen

concentration conditions. Wen et al. (2017a) designed and manufactured the XKGW-1 type high-temperature experimental device to study the influence of the oxygen concentration on the characteristic of coal spontaneous combustion. Wen et al. (2017b) made a forecast model for the concentration of CO at the air return corner under coal mine, which can be used to precisely evaluate the degree of coal spontaneous combustion. Ma et al. (2020) proposed a new index D-O to forecast the coal temperature during the process of coal spontaneous combustion. Zhai et al. (2021) used thermogravimetry to determine characteristic temperatures to judge low-temperature oxidation stages of water-immersion coal.

Coal spontaneous combustion is a highly compound procedure. The characteristics of coal spontaneous combustion are mainly studied from two scales: micro and macro (Wang et al. 2021; Deng et al. 2018; Fan et al. 2021). The particle size is one of the main external factors affecting coal spontaneous combustion (Zhang et al. 2019; Yi et al. 2014a; Yi et al. 2014b). The smaller particle size could facilitate occurrence of coal spontaneous combustion (Fernandez-Anez and Garcia-Torrent 2019), enhancing the oxidizability and exothermicity of coal spontaneous combustion and releasing more gas production (Deng et al. 2017). Zhang et al. (2018) conducted laboratory experiments of coal spontaneous combustion under non-isothermal conditions, and the consequence showed that the total CO output of small particle size coal is more than that of larger particle size coal. Luo et al. (2019) pointed out that, as the particle size decreases, the content of CO, light hydrocarbons, and NO_x precursors all showed a tendency of decrease after the increase during rapid pyrolysis of coal. Rifella et al. (2019) conducted

78 a comprehensive analysis of the crossing-point temperature and critical self-ignition
79 temperature and found that larger particles have a lower risk of spontaneous combustion.
80 The integral characteristic of the low-temperature gas-phase ignition of the products of
81 the thermal decomposition of coal particles is determined (Glushkov et al. 2015). Qin
82 et al. (2012) proposed the oxygen consumption rate equation of coal samples with the
83 mixed particle size, which can be used to accurately calculate the true oxygen
84 consumption rate of coal samples. Jia et al. (2021) found that the smaller particle size
85 of coal improves the adsorption capacity of coal and oxygen molecules, and boosts the
86 oxygen consumption rate.

87 To sum up, the particle size could affect the production of gas during the process of
88 coal-oxygen compound reaction, also affects the accuracy selection of coal spontaneous
89 combustion indexes. To reduce the effects of particle size on predictive index of
90 spontaneous combustion, the coefficient of variation and weighted grey relational
91 analysis were combined to evaluate the accuracy of grading forecast of coal
92 spontaneous combustion index by the temperature-programmed experiments in this
93 study.

2 Experimental Methodology

2.1 Coal samples

The coal sample used in this study was collected from the Lu'an mining area in Shanxi Province, China. Being crushed in a dry environment, the coal samples in the partial size ranges of 0~0.9 mm, 0.9~3 mm, 3~5 mm, 5~7 mm, and 7~10 mm were sieved. A one-kilogram mixed coal sample was prepared by mixing 200 grams from each of the five partial sizes.

2.2 Experimental device

This study employed the temperature-programmed experimental system developed by Xi'an University of Science and Technology. The experimental device consisted of four parts: gas channel, temperature control box, gas collection area, and gas chromatograph. The temperature-programmed experimental system was shown in Fig. 1.

Fig. 1 XK temperature-programmed experimental system

2.3 Experimental process

The coal sample was placed in an experimental steel pipe with a diameter of 10 cm and a length of 22 cm. To ventilate evenly, leave a space of about 2 cm at the upper and

lower ends (using 100 mesh copper wire to fix the coal sample), and then ensure the airtightness of the gas path. The steel pipe was put in the temperature-programmed box and injected to preheated air, followed by the collection of gases generated at different coal temperatures. During the experiment, the heating rate of the coal body was controlled to be 0.3°C/min, and the airflow rate was 120 ml/min. Heating from 30°C to 170°C, the composition and concentration of the collected gases were analyzed by gas chromatography every 10°C during the heating process.

3. Results and discussion

3.1 Characteristic temperature

Critical temperature and dry cracking temperature are two important characteristic temperatures in the process of coal spontaneous combustion. They could be used to judge the degree of coal oxidation, which provides a basis for the early grading forecast of coal spontaneous combustion (Xiao et al. 2017; Hu and Xia 2017). As the oxygen consumption rate is one of the crucial factors affecting coal self-heating capacity (Zhu et al. 2012), it can precisely correspond to the degree of coal spontaneous combustion. Therefore, in this study, the growth rate of oxygen consumption rate with temperature was used to determine the characteristic temperatures of coal spontaneous combustion (Deng et al. 2014b; Zhao et al. 2019). The equation of the growth rate is as follows:

$$B = \frac{c_{i+1} - c_i}{t_{i+1} - t_i} \quad (1)$$

Where c is the oxygen consumption rate, t is the temperature and B is the gradient of oxygen consumption rate as the temperature rises by 1°C.

$$Z = \frac{B}{c_i} \quad (2)$$

Where Z is the growth rate of oxygen consumption rate at a temperature of t_{i+1} .

From Eqs. (1) and (2), Eq. (3) can be obtained:

$$Z = \frac{c_{i+1} - c_i}{c_i(t_{i+1} - t_i)} \quad (3)$$

Fig. 2 shows the curve of oxygen consumption rate with temperature for the coal samples.

Fig. 2 Curve of the oxygen consumption rate with temperature

It can be observed from Fig. 2 that the oxygen consumption rates of coal samples are positively correlated with temperature. As the coal temperature reaches 60°C, the curve has an inflection point, and the slope of the curve increases. Further, as the coal temperature exceeds 80°C, the inclination of the curve becomes gentle. Around 100°C, the curve appears at the inflection point. After 110°C, there is no inflection point on the

curve. Fig. 3 shows the curve of the growth rate of oxygen consumption rate with temperature for the different coal samples.

Fig. 3 The growth rate of oxygen consumption rate with temperature

Fig. 3 indicates that change rules of the growth rate of oxygen consumption rate with temperature for all coal samples are similar. At 60°C~70°C, the first maximum value appears, followed by the second one at 100°C~110°C. It indicates that the oxygen consumption rate elevates rapidly in these temperature ranges. So, 60°C~70°C could be determined to be the critical temperature range for the coal sample, whereas 100°C~110°C is identified as the dry cracking temperature range.

3.2 Single index gas

3.2.1 CO

Fig. 4 shows the variation of CO concentration with temperature for the coal samples.

It can be observed that CO gas is detected at the initial temperature (30°C) for each coal

159 sample. The CO concentrations for all coal sample increase with temperature, and the
 160 influence of particle size on CO production account for its inapplicability for index gas.

161
 162 **Fig. 4** The variation of CO concentration with temperature

163 3.2.2 C_2H_4 and C_2H_6

164 Fig. 5 and 6 show the variation of C_2H_4 and C_2H_6 concentrations with temperature for the
 165 coal samples.

Fig. 5 Curve of the variation of C₂H₄ with temperature

Fig. 6 Curve of the variation of C₂H₆ with temperature

They indicate that the increasing concentration of C₂H₄ and C₂H₆ with temperature are associated with coal particle size. At the same temperature, the smaller particle size coal

sample could generate more C_2H_4 and C_2H_6 gases. Both gases can be detected as the temperature reaches above $110^\circ C$. The generation of C_2H_6 and C_2H_4 gases is related to the high-temperature cracking of coal. Therefore, the appearance of C_2H_4 and C_2H_6 can be used as the index gas to determine the dry cracking temperature.

3.3 Composite index

Due to the complex environment, such as airflow under coal mines, the air would affect the single index gas concentration. It is difficult to precisely judge the degree of coal spontaneous combustion by single gas concentration, while the composite index involving the ratio of at least two gas concentrations can reduce the influence of the environment on the indexes (Liang et al. 2019).

3.3.1 CO_2/CO

Fig. 7 shows the variation of CO_2/CO of all coal samples with temperature.

Fig. 7 The variation of CO_2/CO with temperature

Fig. 7 indicates that there is a negative correlation between the CO_2/CO value and temperature. It exhibits a certain degree of dispersion before 80°C and the curve fluctuation after 80°C is not intuitively shown in the figure. Therefore, it is necessary to further study whether the influence of coal particle size on the variation of CO_2/CO with temperature is within an acceptable range.

3.3.2 $\text{O}_2/(\text{CO}+\text{CO}_2)$

Fig. 8 shows the variation of $\text{O}_2/(\text{CO}+\text{CO}_2)$ of all coal samples with temperature.

Fig. 8 The variation of $\text{O}_2/(\text{CO}+\text{CO}_2)$ with temperature

The value of $\text{O}_2/(\text{CO}+\text{CO}_2)$ decreases as the temperature rises. At 80°C , the curve approaches the inflection point, and the inclination decreases rapidly. At this stage, the coal temperature reaches the critical temperature, and the coal-oxygen reaction takes place speedily. Consequently, an increment of the oxygen consumption rate as well as the rate of production of CO and CO_2 leads to a decrease of the O_2 content and an increase of the CO

and CO₂ contents, and a decreased amplitude of O₂/(CO+CO₂) value increases accordingly.

As the coal temperature exceeds 80°C, the fluctuations are difficult to be judged directly from the curve, thus, it is necessary to further study the influence of coal particle size on the variation of O₂/(CO+CO₂) with temperature.

3.3.3 O₂/CH₄

Fig. 9 shows the variation of O₂/CH₄ of all coal samples with temperature.

Fig. 9 Curve of the variation of O₂/CH₄ with temperature

As observed from Fig. 9, the O₂/CH₄ value is negatively correlated with the temperature.

Below 90°C, the coal particle size have a certain influence on the O₂/CH₄ value, and as the temperature exceeds 90°C, the fluctuations in the curves are not clear.

3.3.4 CO/CH₄

Fig. 10 shows the variation of CO/CH₄ with temperature.

Fig. 10 The variation of CO/CH₄ with temperature

Fig. 10 illustrates that the CO/CH₄ value gradually increases with the increasing temperature. The values of all coal samples exhibit a certain degree of dispersion, when the coal temperature reaches in the range of 120°C~140°C. Thus, the coal particle size influences the variation of CO/CH₄ with temperature.

3.3.5 O₂/CO

Fig. 11 shows the variation of O₂/CO of all coal samples with temperature.

Fig. 11 The variation of O_2/CO with temperature

Fig. 11 indicates that there is a negative correlation between the O_2/CO value and coal temperature for all samples. Before $80^\circ C$, the differences among the index values of each coal sample were various, and it is difficult to observe directly from the figure after $80^\circ C$.

3.3.6 CO_2/O_2

Fig. 12 shows the variation of CO_2/O_2 of all samples with temperature.

Fig. 12 The variation of CO_2/O_2 with temperature

Fig. 12 shows that there is a positive correlation between the CO_2/O_2 value and coal temperature. The particle size of coal influences the variation of CO_2/O_2 with temperature. But, whether the degree of influence is acceptable requires further study.

3.4 Evaluation of forecast indexes for spontaneous combustion

To ascertain the reliability of the different spontaneous combustion indexes numerically, the coefficient of variation and grey relational analysis were used to evaluate the quantitative spontaneous combustion indexes. Combining with the two characteristic temperatures, three stages could be divided for the classification of coal spontaneous combustion.

3.4.1 Coefficient of Variation Analysis

The coefficient of variation (C_v) is a standardized measure of the dispersion degree for the probability distribution. It reflects the degree of difference in the size of a set of data. The

C_v of more than 30% usually indicates that this set of data has a large difference and a high degree of dispersion (Brown and Charles 1998; He and Oyadiji 2001). By the C_v of the coal spontaneous combustion indexes at the different coal particle sizes, the influence level can be obtained. A smaller C_v indicates a lower impact of particle size on the coal spontaneous combustion index. The calculate method of coefficient of variation is as Eq. (4):

$$C_v = \frac{\sqrt{\frac{1}{n-1} \sum_{i=1}^n (X_i - \bar{X})^2}}{\bar{X}} \quad (4)$$

Where C_v is the coefficient of variation, n is the number of particle size, X_i is the index value of spontaneous combustion of all coal samples at the same temperature, and $\bar{X}$ is the average value of X_i .

Figs. 13(a) and (b) show the C_v of coal spontaneous combustion indexes with coal temperature. Fig. 14 shows the average C_v .

(a)

(b)

Fig. 13 The C_v of indexes with coal temperature

Fig. 14 The average C_v of coal spontaneous combustion indexes

Fig. 13 illustrates that the C_v values of CO_2/CO 、 $\text{O}_2/(\text{CO}+\text{CO}_2)$ 、 O_2/CO and CO_2/O_2 at overwhelming majority temperature are less than 20%. Only a small fraction exceeds 20%. Half of the C_v value of CO/CH_4 is below 20%. The C_v value of O_2/CH_4 at each temperature point is higher than 20%, further, the C_v value at a few temperature points is above 30%. Subsequently, Fig. 14 illustrates that the average C_v value for each spontaneous combustion index was determined. Among them, C_v value of O_2/CH_4 is the highest and exceeds 30%, indicating that the dispersion degree of values of O_2/CH_4 of each coal sample is the highest, and the coal particle size has a greater influence on it, other indexes with lower C_v values are less influenced by coal particle size.

3.4.2 Grey relational analysis

The grey relational coefficient could be used to represent the difference degree of the geometric shapes between the curves (Ren et al. 2020; Zhao et al. 2020). The rate of oxygen consumption can be closely associated with the coal sample temperature. Therefore, a

reference series reflecting the system characteristics are determined from the rate of oxygen consumption (If the coal spontaneous combustion indexes show a downward trend with the temperature change, the reciprocal of the rate of oxygen consumption is used as a reference series to reflect the characteristics of the system). The coal spontaneous combustion index values form the comparison series (dimensionless) and average (used in this study) value and initial value methods are usually used to generate the reference series. The corresponding equation is as Eq. (5):

$$\xi_i(k) = \frac{\min_k |t(k) - X_i(k)| + \zeta \max_k |t(k) - X_i(k)|}{|t(k) - X_i(k)| + \zeta \max_k |t(k) - X_i(k)|} \quad (5)$$

Where $\xi_i(k)$ is the grey relational coefficient of X_i to t at time k , ζ is the resolution coefficient (generally between 0 and 1, 0.5 in this study), where $\min_k |t(k) - X_i(k)|$ and $\max_k |t(k) - X_i(k)|$ are the two-level minimum and two-level maximum respectively.

The grey relational coefficient γ_i represents the mean value of $\xi_i(k)$. Fig. 15 shows the grey relational coefficient of each coal spontaneous combustion index. Among them, except for CO/CH₄, the grey correlation coefficients of the other indexes are higher than 0.75, which are at a relatively high level. The grey correlation coefficient of CO/CH₄ is 0.66, which is at a low level.

Fig. 15 The grey relational coefficient of different coal spontaneous combustion indexes

3.4.3 Optimization analysis of weighted grey relation

The influence of particle size on the variation of coal spontaneous combustion indexes with temperature cannot be ignored. In this study, the weighted grey relational analysis is used to optimize the selection of the coal spontaneous combustion index, which can make the selection of prediction and forecast index of coal spontaneous combustion more precise and reliable. The weighted of the C_v is as Eq. (6):

$$w_i = \frac{1}{C_v} \quad (6)$$

Where w_i is the weighted coefficient of variation.

The equation for the weighted grey relational degree can be given as Eq. (7):

$$R_i = \frac{1}{n} \sum_{i=1}^n \zeta_i(K) \cdot w_i \quad (7)$$

Fig. 16 shows the weighted grey relational degree of various indexes and their ranking results. It can be seen that $R_{CO_2/CO}$ is the highest, followed by R_{CO_2/O_2} and $R_{O_2/(CO+CO_2)}$, and

304 R_{CO/CH_4} and R_{O_2/CH_4} are at a lower level, indicating that the reliability of CO_2/CO is the
305 highest as a coal spontaneous combustion index. As the limit value, the median R_{median} was
306 used to select the indexes. When the weighted grey relational degree is higher than the
307 R_{median} , it could be the prediction and forecast index of coal spontaneous combustion.
308 Therefore, it is recommended to select CO_2/CO as the main index, and CO_2/O_2 and
309 $O_2/(CO+CO_2)$ as the auxiliary index.

311 **Fig. 16** The weighted grey relational degree of each coal spontaneous combustion indexes

312 The grading forecasting index system of coal spontaneous combustion is shown in Table 1.

313 **Table 1** Grading forecasting index system of coal spontaneous combustion

Index type	First index	Second index		Confirmation index	
Index content	CO_2/CO	CO_2/O_2	$O_2/(CO+CO_2)$	C_2H_4	C_2H_6
Critical temperature /60~70°C	20.68~39.62	0.006~0.013	74.54~154.92		
Dry cracking temperature /100~110°C	4.18~6.52	0.056~0.135	6.18~15.03	appears	appears

4. Conclusions

In this study, the growth rate analysis method was used to identify characteristic temperatures of the coal spontaneous combustion by temperature-programmed test. the critical temperature range for the coal sample is determined to be 60°C~70°C, whereas the dry cracking temperature range is 100°C~110°C. Based on the variation of CO, C₂H₄, and C₂H₆ during coal oxidation with consideration of particle sizes, C₂H₄ and C₂H₆ were selected as the confirmation indexes for the coal temperature to reach the dry cracking temperature.

The coefficient of variation was used to analyze the influence of coal particle size on composite indexes, CO₂/CO, CO₂/O₂, O₂/(CO+CO₂), O₂/CO, CO/CH₄, and O₂/CH₄. By weighted grey relational analysis, the quantitative coal spontaneous combustion indexes were proposed, CO₂/CO as the first index, CO₂/O₂, and O₂/(CO+CO₂) as the second index.

Statements and Declarations

CRediT authorship contribution statement

Yang Li: Conceptualization, Methodology, Writing-original draft. Kai Wang: Validation, Writing-review & editing. Yanni Zhang: Writing-review & editing. Jun Deng: Writing-review & editing.

Ethical Approval

Not applicable.

Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Consent to Participate

Not applicable.

Consent to Publish

The Author agrees to publication in “*Environmental Science and Pollution Research*” and also to publication of the article in English by Springer in Springer's corresponding English-language journal.

Availability of data and materials

The datasets used or analysed during the current study are available from the corresponding

351 author on reasonable request.

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