Improved Calculation Model for the Shortest Spontaneous Combustion Period

Yulong Gao, Siyi Lin, Wenhua Hu, and Shuping Yi*

ABSTRACT: Disasters caused by the spontaneous combustion of coal have occurred in major coal-producing countries, resulting in the loss of resources and human life and severe environmental pollution. The development of an efficient model to calculate the shortest spontaneous combustion period (SSCP) has been a longstanding challenge. In this study, we propose a continuous model that calculates the SSCP by changing the traditional time summation form into a time integration form. The proposed model can reduce the calculation errors and determine the heating time to any temperature, which overcomes the limitations of the traditional model. The accuracy and convenience of the improved model were validated through a comparison with the results of the traditional model and a numerical model. The parameter sensitivities were analyzed in the improved model. The results showed that the SSCP calculated using the improved continuous model are in good agreement with those of the traditional and numerical models. The results also indicated that the continuous model is more accurate and convenient than the traditional model. Parameters such as the heat release intensity, water content, specific heat capacity, and gas content influence the SSCP results in the sensitivity analysis. This model can potentially help prevent and control the risk of coal spontaneous combustion and should be further tested in practical mine management.

1. INTRODUCTION

Coal is one of the predominant energy sources in many countries, and 5.5 billion tons of coal were consumed globally in 2015, accounting for more than 19% of the world’s primary energy consumption.1−3 From 2007 to 2017, coal consumption in Europe and North America declined, while consumption in Asia continued to grow.4 However, some coal seams are prone to spontaneous combustion. Coal spontaneous combustion disasters have occurred in the USA, India, Australia, Indonesia, Poland, South Africa, China, and other major coal-producing countries.5−6 Long-term coal storage at thermal power plants, surface coal mining spoil piles, and long-distance transportation in cargo ships or trains may cause self-heating of the coal stockpile.6 The spontaneous combustion and smoldering of coal stockpiles pose significant threats to coal producers and users.9−11 In addition, severe and extensive oxidation of coal can also cause loss of its calorific value and coking property.11−13 Moreover, heat continues to accumulate during the accumulation process, which releases high concentrations of SO2, NOx, CO, and other harmful gases into the atmosphere, leading to severe environmental problems.14−18 Underground coal combustion has also led to surface subsidence and the subsequent occurrence of geological disasters.19,20 As a result, studies on the spontaneous combustion of coal remain important global tasks.

Coal of all ranks can be oxidized at low temperature. It is widely recognized that low-temperature oxidation is the main source of heat leading to the spontaneous combustion of coal. The low-temperature interactions between coal and oxygen are generally exothermic, although some reactions may be endothermic.21 Hazards develop when the heat release rate is greater than the cooling rate. Owing to the poor thermal conductivity of coal, excess heat is stored in the coal, which leads to an increased temperature and accelerated oxidation rate. Once the temperature of the coal exceeds the critical temperature, a fire will soon occur if appropriate remedial measures are not enacted.22 However, once active heating is detected, it may already be too late to take action. The established common wisdom in dealing with spontaneous combustion is thus that prevention is always better than a cure. As a result, studies on the spontaneous combustion of coal must be used to infer the spontaneous combustion development process, allowing effective fire prevention measures to be employed in time to avoid the loss of resources or life and to mitigate environmental pollution.

Many studies on forecasting the spontaneous combustion of coal have been carried out, including field monitoring of coal mines, direct tests based on model experiments, numerical
models, and analytical models. In field monitoring, temperature or index gas probes are placed in the field, which allows the spontaneous combustion of coal in a mine to be effectively managed. However, field monitoring is expensive, and it is difficult to cover the entire area of concern. As a result, coal spontaneous combustion may not be predicted sufficiently early for prevention. To predict the spontaneous combustion locations and ignition period of coal more accurately, the model experiment on spontaneous combustion has been developed with a higher degree of simulation and more accurate and reliable data, which has successfully solved the key problems of on-site simulation of coal spontaneous combustion.\textsuperscript{33–25} However, this method is more time-consuming and has a higher cost and poorer repeatability. The numerical model was established based on the actual environment in the field to forecast the spontaneous combustion process by data-computing programs.\textsuperscript{7,26–29} However, the applicability of the model is poor, and the results are uncertain and difficult to verify owing to the complexity of actual environmental conditions.

Analytical models use parameters tested in laboratory settings based on the heat balance relationship to obtain the shortest spontaneous combustion period (SSCP). Results can be obtained rapidly and with low cost. Some analytical models have also been developed. Boddington\textsuperscript{30,31} proposed an expression to calculate the time to ignition based on kinetic parameters (the activation energy, \( E \), and pre-exponential factor, \( A \)), but neglected the variation in these kinetic parameters caused by an increase in the temperature. Crrien\textsuperscript{32} developed a model based on the heating of coal caused only by the exothermic absorption of oxygen, whereas the exothermic oxidation (the main source of heat in the self-heating process) was ignored. Yu\textsuperscript{33} modified the aforementioned model by including oxidation heat; the variation in parameters caused by temperature was also considered. The mean values of parameters such as the specific heat capacity and oxidative heat release intensity are calculated at different temperature intervals, for example, 10–20, 30–40, and 40–50 °C in Yu’s model. Therefore, the calculation process is discontinuous. Yang\textsuperscript{34} performed a similar modification to that of Yu’s model. In these traditional analytical models, the heating time must be divided into different temperature intervals in the calculation process owing to the variation in the parameters, specific heat capacity, heat release intensity, water content, and gas content with temperature. The parameter values are averaged for each temperature interval, which can cause errors. To minimize these errors, the temperature interval should be as small as possible; however, this increases the workload substantially. In addition, these models can only calculate the heating time to temperatures at the ends of these intervals and is not able to calculate the heating time to temperatures that fall within these intervals. Therefore, it is necessary to develop a model to calculate the SSCP more accurately and conveniently.

This study establishes a continuous model that can reduce the errors of the traditional model. Another advantage of the improved model is that it can be used to calculate the heating time to any temperature, which is more convenient than the traditional model. The accuracy and convenience of the improved model are verified by comparison to traditional analytical and numerical models. In addition, the sensitivity of parameters is analyzed based on the continuous model, and the influence of the parameters is confirmed.

2. METHODS

2.1. Model Derivation. The actual environment can be considered in an adiabatic state during coal spontaneous combustion owing to the small amounts of coal thermal loss and air leakage.\textsuperscript{33} The exothermic oxidation of coal is used for coal heating, water evaporation, and gas desorption. The heat balance equation for loose coal is as follows\textsuperscript{34}

\[
Q_c = \frac{\rho C \Delta T}{\Delta t} + Q_g + Q_w
\]

where \( Q_c \) is the oxidation exothermic rate, J/(m\(^3\) s); \( \rho \) is the density of loose coal, kg/m\(^3\); \( C \) is the specific heat capacity of coal, J/(kg·K); \( \Delta T \) is the temperature change, \( s \); \( Q_g \) is the endothermic gas desorption rate, J/(m\(^3\) s); and \( Q_w \) is the moisture evaporation heat rate, J/(m\(^3\) s).

The time change can be expressed as follows based on eq 1

\[
\Delta t = \frac{\rho C \Delta T + q_g \Delta X_g + q_w \Delta M_w}{Q_c}
\]

where \( X_g \) is the gas desorption capacity, m\(^3\)/kg; \( q_g \) is the heat of gas desorption, 1.26 \times 10\(^7\) J/m\(^3\); \( M_w \) is the moisture evaporation, kg/m\(^3\); and \( q_w \) is the heat of moisture evaporation, 2.26 \times 10\(^7\) J/kg.

The parameters, specific heat capacity, heat release intensity, water content, and gas content vary with temperature. Thus, the heating time needs to be divided into different temperature intervals in the calculation process. Then, the SSCP is expressed as the sum of all time intervals as follows

\[
\tau = \sum \left( \frac{\rho C \Delta T + q_g \Delta X_g + q_w \Delta M_w}{Q_c} \right)
\]

The above equation describes the traditional model for calculating the SSCP. The values of the parameters in eq 3 are replaced by averages in each temperature interval, which can cause errors. To reduce this error, smaller temperature intervals must be used. However, this increases the workload of the calculation model, and the errors still exist. In addition, the traditional model can only calculate the heating time to the dividing point of the temperature ranges; it cannot calculate the heating time to any given temperature. To improve the traditional model, we changed the summation form of the traditional model to an integral form. The parameters, specific heat capacity \( C \), heat release intensity \( Q \), water content \( M_w \), and gas content \( X_g \), which vary with temperature, are considered as functions of temperature. These parameters can be written as \( C(T) \), \( Q(T) \), \( X_g(T) \), and \( M_w(T) \). The forms of \( X_g(T) \) and \( M_w(T) \) cannot be used directly, and thus need to be further modified in the new model. Based on the Taylor formula and neglecting higher-order terms, \( X_g(T) \) and \( M_w(T) \) can be written as follows

\[
X_g(T + \Delta T) = X_g(T) + X_g'(T) \Delta T
\]

\[
M_w(T + \Delta T) = M_w(T) + M_w'(T) \Delta T
\]

By moving the first terms on the right side of the equations to the left side, eqs 4 and 5 can be written as

\[
X_g(T + \Delta T) - X_g(T) = X_g'(T) \Delta T
\]

\[
M_w(T + \Delta T) - M_w(T) = M_w'(T) \Delta T
\]
\( \Delta X(T) \) and \( \Delta M_c(T) \) replace the terms on the left sides of eqs 6 and 7, respectively. The original equations can thus be written as

\[
\Delta X(T) = X(T) \Delta T
\]

\( \Delta M_c(T) = M_c(T) \Delta T
\)

The other parameters, obtained in the next section.

Through theoretical derivation, the summation form has been avoided. In addition, the new model can calculate the heating temperature for the coal samples.

2.2. Model Parameter Processing. In the improved SSCP model, the specific heat capacity, heat release intensity of oxidation, and gas desorption under different temperatures are needed. Six coal samples were selected from Yujialiang, Ciyaowan, Shangwan, Zhangji, Yangzhuang, and Wushi, where spontaneous combustion events have occurred in China. The sample parameters were measured at different temperatures and fitted using different function models. Table 1 presents the industrial analysis results for the samples.

### Table 1. Industrial Analysis of Coal Samples

| Coal sample | Ash (%) | Moisture (%) | Volatiles (%) | Fixed Carbon (%) | Density (kg/m³) |
|-------------|---------|--------------|---------------|-----------------|----------------|
| Yujialiang  | 7.4     | 6.98         | 31.42         | 54.19           | 1263           |
| Ciyaowan    | 6.09    | 8.99         | 32.65         | 52.27           | 1265           |
| Shangwan    | 6.7     | 9.21         | 27.86         | 56.23           | 1237           |
| Zhangji     | 8.18    | 2.07         | 20.75         | 80.21           | 1191           |
| Yangzhuang  | 13.84   | 0.88         | 15.42         | 90.44           | 1295           |
| Wushi       | 19.09   | 0.8          | 26.18         | 87.95           | 1357           |

2.3. Moisture Evaporation. The moisture evaporation can be determined from the thermogravimetric (TG) curve obtained using thermogravimetry. Thermogravimetry is a technique to measure the relationship between the mass and temperature of a substance under varying temperatures controlled by the program. Fresh coal samples were stripped of the outer coal layer and ground before testing. The experiments were carried out under a nitrogen environment. More details regarding the experimental processes can be found in Yang et al.\(^{35}\) The TG data were from Yang et al.\(^{35}\)

The moisture evaporation can be calculated based on the TG data as follows

\[
M_w = \rho (1 - TG)
\]

The moisture evaporation of samples is calculated according to eq 12, and the relationship between the moisture evaporation and temperature is shown in Figure 1.

As seen in Figure 1, the moisture evaporation \( M_w \) increases and becomes almost stable with increasing temperature, which indicates that the rate of evaporation decreases gradually. An exponential function is thus used to fit the moisture evaporation data in this study. The expressions for \( M_w \) are given as follows

\[
M_{w1} = -466.61 e^{(T-11.34)/27.35} + 80.92
\]

\[
M_{w2} = -193.22 e^{(T-16.93)/20.47} + 58.93
\]

\[
M_{w3} = -272.03 e^{(T-24.69)/24.69} + 115.69
\]

\[
M_{w4} = -74.23 e^{(T-30.43)/23.56} + 38.13
\]

\[
M_{w5} = -44.50 e^{(T-27.35)/24.69} + 20.47
\]

\[
M_{w6} = -63.03 e^{(T-28.55)/28.39}
\]

where \( M_{w1}, M_{w2}, M_{w3}, M_{w4}, M_{w5}, \) and \( M_{w6} \) are the moisture evaporation of the Yujialiang, Ciyaowan, Shangwan, Zhangji, Yangzhuang, and Wushi coal samples, respectively (in kg/m³).

The mean relative error (MRE) is used to evaluate the effectiveness of the fitting in this study. The MRE is calculated as follows

\[
\text{MRE} = \frac{1}{n} \sum_{i=1}^{n} \left| \frac{P_i - O_i}{O_i} \right|
\]

The MRE results are listed in Table 2. The MRE values of all samples are less than 7%, which indicates that the fitting of the moisture evaporation data using eq 13 is feasible.

Then, the expression for \( M_w \) required for the new continuous model can be calculated with eq 15.
between the heat release intensity and the temperature can also considerably with increasing temperature. The relationship between the experimental processes are provided in Yang et al.\textsuperscript{37} Figure 2. Relationship between the heat release intensity and the temperature. Adapted with permission from ref\textsuperscript{34}. Copyright 2014, Elsevier.

As seen in Figure 2, the heat release intensity ($Q_c$) increases considerably with increasing temperature. The relationship between the heat release intensity and the temperature can also be fitted with an exponential function. A single non-piecewise exponential function is used to fit the heat release intensity data. The expressions for $Q_c$ are given as follows

$$Q_{c1} = 1.61 e^{(7/11.27)} + 36.76$$
$$Q_{c2} = 4.03 e^{(7/17.54)} + 34.49$$
$$Q_{c3} = 4.01 e^{(7/17.62)} + 96.92$$
$$Q_{c4} = 7.73 e^{(7/11.09)} - 34.47$$
$$Q_{c5} = 0.92 e^{(7/15.44)} - 10.48$$
$$Q_{c6} = 9.53 e^{(7/22.83)} - 42.18$$

(15)

$M_{w1}, M_{w2}, M_{w3}, M_{w4}, M_{w5}$, and $M_{w6}$ are the derivatives of $Q_{c1}, Q_{c2}, Q_{c3}, Q_{c4}$, $Q_{c5}$, and $Q_{c6}$ respectively.

2.4. Heat Release Intensity. The heat release intensity of the coal samples was tested through loose coal oxidation experiments, which is a new method based on heat conduction theory.\textsuperscript{36,37} A measured amount of coal was placed into a coal oxidation test system. The system can collect data regarding the temperature and oxygen content in the process of coal oxidation. The heat release intensity of the coal samples was obtained by solving the calculation model based on the collected experimental data. More details of this method and the experimental processes are provided in Yang et al.\textsuperscript{37} Figure 2 shows the relationship between the heat release intensity and the temperature.

![Figure 2. Relationship between the heat release intensity and temperature. Adapted with permission from ref 34. Copyright 2014, Elsevier.](image)

### Table 2. MREs of the Fitted Moisture Evaporation Data

| coal sample     | Yujialiang | Ciyaowan | Shangwan | Zhangji | Yangzhuang | Wushi |
|-----------------|------------|----------|----------|---------|------------|-------|
| MRE (%)         | 1.98       | 1.04     | 6.17     | 1.17    | 6.15       | 5.96  |

### Table 3. MREs of the Fitted Heat Release Intensities

| coal sample     | Yujialiang | Ciyaowan | Shangwan | Zhangji | Yangzhuang | Wushi |
|-----------------|------------|----------|----------|---------|------------|-------|
| MRE, eq 16 (%)  | 17.11      | 2.42     | 7.57     | 29.39   | 37.74      | 37.74 |
| MRE, eq 17 (%)  | 1.98       | 1.04     | 6.17     | 1.17    | 6.15       | 5.96  |

The MREs between the heat release intensities fitted using eq 16 and the corresponding experimental values are given in the first row of Table 3. As indicated in Table 3, the MREs for the Yangzhuang and Wushi samples are large (greater than 30%), which is unacceptable. The heat release intensity values differ considerably at low and high temperatures (Figure 2), which leads to large fitting errors. To reduce the relative error, the heat release intensities are fitted separately at low temperatures and high temperatures. Thus, 70 °C is set as the demarcation point of the piecewise function. The piecewise exponential function bounded by 70 °C is given as follows

$$Q_{c1} = 0.06 e^{(7/12.27)} + 26.60 \quad 20 ^\circ C \leq T < 70 ^\circ C$$
$$Q_{c2} = 0.96 e^{(7/14.90)} + 108.11 \quad T \geq 70 ^\circ C$$
$$Q_{c3} = 9.54 e^{(7/21.76)} + 15.61 \quad 20 ^\circ C \leq T < 70 ^\circ C$$
$$Q_{c4} = 3.92 e^{(7/17.49)} - 38.78 \quad T \geq 70 ^\circ C$$
$$Q_{c5} = 26.71 e^{(7/28.55)} + 34.38 \quad 20 ^\circ C \leq T < 70 ^\circ C$$
$$Q_{c6} = 4.92 e^{(7/17.54)} + 51.57 \quad T \geq 70 ^\circ C$$
$$Q_{c7} = 0.15 e^{(7/10.47)} + 26.40 \quad 20 ^\circ C \leq T < 70 ^\circ C$$
$$Q_{c8} = 21.32 e^{(7/25.46)} - 201.43 \quad T \geq 70 ^\circ C$$
$$Q_{c9} = 0.20 e^{(7/11.90)} + 18.61 \quad 20 ^\circ C \leq T < 70 ^\circ C$$
$$Q_{c10} = 1.04 e^{(7/15.89)} + 0.91 \quad T \geq 70 ^\circ C$$
$$Q_{c11} = 0.10 e^{(7/10.19)} + 24.65 \quad 20 ^\circ C \leq T < 70 ^\circ C$$
$$Q_{c12} = 38.19 e^{(7/30.61)} - 269.58 \quad T \geq 70 ^\circ C$$

(17)
The MREs between the heat release intensities fitted using eq 17 and the experimental values are given in the second row of Table 3. The MRE values for all samples are less than 7%, which suggests that fitting the heat release intensity data using eq 17 is feasible.

### 2.5. Specific Heat Capacity

The specific heat capacity of the coal samples was also tested through loose coal oxidation experiments, which are similar to the tests for the heat release intensity. A measured amount of coal was placed into a coal oxidation test system that could collect the temperature and oxygen content data during the coal oxidation process. The specific heat capacity of the coal samples was obtained based on the data collected in these experiments. Yang et al. describe the relevant method and experimental processes. Figure 3 shows the relationship between the specific heat capacity and the temperature.

![Figure 3](image-url)

Figure 3. Relationship between the specific heat capacity of coal samples and the temperature. Adapted with permission from ref 34. Copyright 2014, Elsevier.

As Figure 3 shows, the specific heat capacity (C) of the coal samples increases linearly with increasing temperature. Thus, a linear function is used to fit the specific heat capacity data in this study. The expressions for C are given as follows

\[ C_1 = 2.82T + 1087.62 \]
\[ C_2 = 3.71T + 1091.82 \]
\[ C_3 = 3.99T + 981.41 \]
\[ C_4 = 4.61T + 732.43 \]
\[ C_5 = 1.87T + 799.41 \]
\[ C_6 = 2.44T + 871.68 \]

(18) where \( C_{1} \), \( C_{2} \), \( C_{3} \), \( C_{4} \), \( C_{5} \), and \( C_{6} \) are the specific heat capacities of the Yujialiang, Ciyaowan, Shangwan, Zhangji, Yangzhuang, and Wushi coal samples, respectively [in J/(kg·K)].

The MREs of the fitted specific heat capacity values are listed in Table 4. The MREs of all samples are less than 2%, which suggests that fitting the specific heat capacity data using eq 18 is feasible.

### 2.6. Gas Desorption

When coal is exposed to the atmosphere, it is assumed that the gas inside the coal will be released to the atmosphere. The gas content can thus be calculated using the Langmuir equation

\[ V_s = \frac{(1 - A - M) abp}{1 + bp} \]

(19) where \( V_s \) is the gas content, m³/kg; \( A \) is the ash percentage, 1; \( M \) is the moisture content percentage, 1; \( a \) is an adsorption constant, m³/kg; \( b \) is an adsorption constant, Pa⁻¹; and \( p \) is the gas pressure, Pa.

Parameters \( a \) and \( b \) can be calculated according to the ash or volatile contents. The results for parameters \( a \) and \( b \) are listed in Table 5.

It has been demonstrated that 20% of the adsorbed gas is released from room temperature to 60 °C, 40% of the gas is desorbed from 60 to 100 °C, and the remainder is desorbed from 100 to 160 °C. The gas desorption can thus be approximated by the average processes at different temperature stages. The gas desorption capacity, \( X \), can be expressed as follows

\[ X = \begin{cases} \frac{V_s T - 20}{200} & 20°C \leq T < 60°C \\ \frac{V_s T - 60}{100} & 60°C \leq T < 100°C \\ \frac{V_s T - 100}{150} & 100°C \leq T < 160°C \end{cases} \]

(20)

Then, the derivative of \( X \) (\( X' \)), which is needed for the continuous model, can be expressed as follows

\[ X' = \begin{cases} \frac{V_s T}{200} & 20°C \leq T < 60°C \\ \frac{V_s T}{100} & 60°C \leq T < 100°C \\ \frac{V_s T}{150} & 100°C \leq T < 160°C \end{cases} \]

(21)

### 3. RESULTS AND DISCUSSION

#### 3.1. Comparison with the Traditional Analytical Model

An improved continuous model was developed in Section 2. In this section, the accuracy of the continuous model is verified by comparing the SSCP’s calculated with the continuous model to those obtained using the traditional discontinuous model. The SSCP’s were calculated with the continuous model based on eq 11, while calculation in the traditional model was based on eq 3. For the traditional discontinuous model, it is necessary to determine the temperature interval (\( \nabla T \)). Within each \( \nabla T \), the parameters are averaged. In this section, the heating time of the traditional discontinuous model is calculated at temperature intervals (\( \nabla T \)) of 30, 20, and 10 °C. The heating times calculated by the continuous model are compared to those obtained using the traditional model. Figure 4 shows the heating times for six coal samples calculated using the continuous model and the traditional model with

| coal sample     | Yujialiang | Ciyaowan | Shangwan | Zhangji | Yangzhuang | Wushi |
|-----------------|------------|----------|----------|---------|------------|------|
| MRE (%)         | 0.19       | 0.88     | 1.10     | 1.48    | 1.13       | 1.47 |
temperature intervals of 10, 20, and 30 °C. As the temperature interval increases, the calculated heating time of the coal decreases. The heating time for an interval of 10 °C is much lower than those for intervals of 20 or 30 °C, which indicates that overly large temperature intervals will affect the accuracy of the calculation results in the traditional model. Therefore, \( \nabla T \) has a significant influence on the results of the traditional model. Reducing the temperature interval would provide more accurate results, but it will also increase the workload. It can be seen in Figure 4 that the trends in the heating times calculated by the two models are consistent. The heating time calculated using the continuous model is closer to the results calculated using the traditional model with a temperature interval of 10 °C, which has less error than the results calculated at intervals of 20 and 30 °C. Thus, the continuous model is accurate. In addition, the traditional model cannot calculate the heating time to any given temperature. For example, when \( \nabla T \) is 30 °C, the traditional model can only calculate the heating time from 20 to 50 °C, 80, 110 °C, and so forth. In contrast, the new model can calculate the heating time to any temperature of interest. Therefore, the heating time can be directly calculated with the continuous model conveniently and with reasonable accuracy.

It can be seen in Figure 4 that the heating time to 70 °C accounts for the main part of the total time. This indicates that 70 °C is a threshold value in the self-heating. The temperature of coal will increase rapidly and spontaneous combustion will soon occur when the temperature exceeds the threshold value. Therefore, it is important to implement prevention and control measures to mitigate the coal spontaneous combustion risk in the previous stage. These results also confirm that it is reasonable to use 70 °C as the dividing point for fitting the heat release intensity in Section 2.4.

3.2. Comparison with a Numerical Model. The results of the continuous model were compared with those obtained using a numerical model to further verify the practicability and accuracy of the proposed continuous model. A two-dimensional trapezoidal coal stockpile is assumed in this section; the bottom length of the coal pile is 6 m, the topline length is 2 m, and the height is 1.5 m. The size of the coal stockpile is shown in Figure 5.

The governing equation of the numerical model is as follows:

Table 5. Values of Parameters \( a \) and \( b \)

| coal sample   | Yujialiang | Ciyaowan | Shangwan | Zhangji | Yangzhuang | Wushi |
|---------------|------------|----------|----------|---------|------------|------|
| \( a \) \((m^3/t)\) | 45.65      | 47.41    | 42.89    | 39.75   | 28.26      | 34.97|
| \( b \) \((1/MPa)\) | 0.29       | 0.22     | 0.37     | 0.49    | 0.91       | 0.72 |

Figure 4. Comparison of heating times calculated using the continuous model and traditional model.
desorption. These heat sources can be calculated with eqs 13, 17 and 20, respectively. The specific heat capacity can be calculated using eq 18.

The initial temperature in the model is 20 °C. The two waists and the upper line of the coal pile are in contact with the air. These sides transfer heat to the air to reduce the risk of spontaneous combustion. The shortest SSCP is the spontaneous combustion time under the maximum risk. Thus, the spontaneous combustion time calculated with those obtained using the continuous model, the numerical model, and the traditional model. The abscissa of a point represents the SSCP calculated using the continuous model (solid points) and the traditional model (open points).

The heating times calculated with the continuous model are compared to those obtained using the numerical model and the traditional model. Figure 6 shows a comparison between the SSCP values (heating time from 20 to 70 °C) calculated using the numerical model, continuous model, and traditional model. In this section, the temperature interval in the traditional model is 10 °C. The SSCP results calculated with the continuous model and the traditional model are consistent. The SSCP values for the Yangzhuang and Wushi coals calculated using the numerical model, continuous model, and traditional model. The SSCP values for the two samples are closer to the results of the numerical model than the traditional model, which indicates that the continuous model is more accurate than the traditional model. Figure 7 shows the heating times from 20 to 30 °C, 40, 50, 60, and 70 °C calculated using the numerical model, the continuous model, and the traditional model. All of the points fall near the dotted line, indicating that the heating times calculated using the numerical model and the traditional model are close to those calculated using the continuous model. The R-squared value can be used to estimate the degree of linear relationship between two variables. Table 6 lists the R-squared values between the heating times calculated with the numerical model and those obtained using the continuous model and the traditional model. The R-squared values between the numerical model and the continuous model and those obtained using the continuous model and the traditional model indicate that the heating times calculated using the continuous model have more stronger linear relationship with those calculated using the numerical model than the traditional model. This also demonstrates that the continuous model is more accurate than the traditional model.

3.3. Sensitivity Analysis. The role of the parameters can be better understood by performing a sensitivity analysis. The relative sensitivity index (RSI) can be used to compare the sensitivity to changes in different parameters; it is expressed as follows:

\[
\text{RSI} = \frac{(\text{SSCP}(\alpha_i + \Delta \alpha_i) - \text{SSCP}(\alpha_i))/\text{SSCP}(\alpha_i)}{\Delta \alpha_i/\alpha_i}
\]

where RSI is the RSI of a parameter; \(\alpha_i\) is the base value of parameter \(i\); \(\Delta \alpha_i\) is the change in parameter \(i\); \(\text{SSCP}(\alpha_i)\) is the value of the SSCP when parameter \(i\) is equal to \(\alpha_i\); and SSCP...
(α + △α) is the value of the SSCP when parameter i is equal to α + △α.

The RSIs of four parameters in the continuous model, that is, the heat release intensity, specific heat capacity, gas content, and water content, are calculated in this section. To calculate the RSI of each parameter, the parameter value is adjusted by 10%, which means △α is equal to 10% of α in eq 23. To compare the sensitivity more intuitively, the absolute value of the RSI is considered. The RSI results for the six coal samples are shown in Figure 8. For the coal samples, the influence of the parameters on the spontaneous ignition period is similar. The heat release intensity has the greatest influence on the spontaneous combustion ignition period. The water content has the second-greatest influence on the spontaneous combustion ignition period in all of the coal samples except that from Yangzhuang. The effect of the specific heat capacity on the spontaneous combustion is the third-most significant for all of the coal samples except that from Yangzhuang. The maximum RSI of the gas content is 0.003 among the six coal samples, which is far lower than the RSIs of the other three parameters. Thus, the gas content has little effect on the SSCP. Among the above parameters, the water content is the easiest to change and has a significant influence on the spontaneous combustion of coal. Therefore, preventing spontaneous combustion of coal can be achieved by changing the water content of coal.

In this section, only the four parameters in the improved model are analyzed. Other factors, such as the dynamic oxygen absorption and activation energy, also have an important influence on coal spontaneous combustion. These parameters are not included in the new model and are not analyzed further.

### 4. CONCLUSIONS

This study established a continuous SSCP model by improving the traditional SSCP model. The model derivation is discussed in detail. The advantages of this model are that it can reduce the error found in the traditional model and calculate the heating time to any given temperature. The accuracy is verified by comparison with results obtained using the traditional discontinuous model and a numerical model. The inconsistencies between the traditional analytical model and numerical model verifies the accuracy of the proposed continuous model. It also shows that the continuous model is more convenient for calculating the heating time to any given temperature than the traditional model. The calculation results for the SSCP indicated that the heating time to 70 °C accounts for the major part of the total time. When the coal temperature exceeds 70 °C, spontaneous combustion of coal will soon occur. Therefore, prevention and control measures to mitigate the risk of the spontaneous combustion of coal should be implemented in the previous stage. Furthermore, the sensitivity of the proposed model is also analyzed. Through parameter sensitivity analyses, their influence on the SSCP results can be ranked in decreasing order of importance as heat release intensity > water content > specific heat capacity > gas content. Considering the effectiveness and maneuverability of prevention measures, changing the water content is a feasible method to prevent the occurrence of coal spontaneous combustion.

### AUTHOR INFORMATION

**Corresponding Author**

**Shuping Yi** — School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, China; State Environmental Protection Key Laboratory of Integrated Surface Water-Groundwater Pollution Control, Shenzhen 518055, China; orcid.org/0000-0003-0136-2321; Email: yisp@sustech.edu.cn

**Authors**

**Yulong Gao** — School of Environment, Harbin Institute of Technology, Harbin 150090, China; School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, China; State Environmental Protection Key Laboratory of Integrated Surface Water-Groundwater Pollution Control, Shenzhen 518055, China

**Siyi Lin** — School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, China; State Environmental Protection Key Laboratory of Integrated Surface Water-Groundwater Pollution Control, Shenzhen 518055, China

**Wenhua Hu** — School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, China; State Environmental Protection Key Laboratory of Integrated Surface Water-Groundwater Pollution Control, Shenzhen 518055, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.0c01831

**Notes**

The authors declare no competing financial interest.
ACKNOWLEDGMENTS

This work was supported partially by the Natural Science Foundation of China (grant no. 41877193), the National Science and Technology Major Project on Water Pollution Control and Treatment (no. 2018ZX07109-002), and the State Environmental Protection Key Laboratory of Integrated Surface Water-Groundwater Pollution Control.

REFERENCES

(1) Li, J.; Wang, J. Comprehensive utilization and environmental risks of coal gangue: A review. J. Clean. Prod. 2019, 239, 117946.

(2) Zhang, Y.; Zhang, Y.; Li, Y.; Li, Q.; Zhang, J.; Yang, C. Study on the characteristics of coal spontaneous combustion during the development and decaying processes. Process Saf. Environ. Prot. 2020, 138, 9–17.

(3) Qiao, L.; Deng, C.; Dai, F.; Fan, Y. Experimental Study on a Metal-Chelating Agent Inhibiting Spontaneous Combustion of Coal. Energy Fuels 2019, 33, 9232–9240.

(4) British Petroleum. BP statistical review of world energy; British Petroleum, 2017.

(5) Song, Z.; Kuenzer, C. Coal fires in China over the last decade: a comprehensive review. Int. J. Coal Geol. 2014, 133, 72–99.

(6) Li, Z.; Kong, B.; Wei, A.; Yang, Y.; Zhou, Y.; Zhang, L. Free radical reaction characteristics of coal low-temperature oxidation and its inhibition method. Environ. Sci. Pollut. Res. 2016, 23, 23593–23605.

(7) Zhu, H.-q.; Song, Z.-y.; Tan, B.; Hao, Y.-z. Numerical investigation and theoretical prediction of self-ignition characteristics of coarse coal stockpiles. J. Loss Prev. Process Ind. 2013, 26, 236–244.

(8) Carras, J. N.; Young, B. C. Self-heating of coal and related materials: models, application and test methods. Prog. Energy Combust. Sci. 1994, 20, 1–15.

(9) Cheng, W.; Hu, X.; Xie, J.; Zhao, Y. An intelligent gel designed to control the spontaneous combustion of coal: fire prevention and extinguishing properties. Fuel 2017, 210, 826–835.

(10) Onifade, M.; Genc, B.; Carpede, A. A new apparatus to establish the spontaneous combustion propensity of coals and coal-shales. Int. J. Coal Sci. Technol. 2018, 28, 649–655.

(11) Swann, P. D.; Allardice, D. J.; Evans, D. G. Low-temperature oxidation of brown coal. I. Changes in internal surface due to oxidation. Fuel 1974, 53, 85–87.

(12) Yang, Y.; Li, Z.; Tang, Y.; Liu, Z.; Ji, H. Fine coal covering for preventing spontaneous combustion of coal pile. Nat. Hazards 2014, 74, 603–622.

(13) Arisoy, A.; Akgüz, F. Effect of pile height on spontaneous heating of coal stockpiles. Combust. Sci. Technol. 2000, 153, 157–168.

(14) Carras, J. N.; Day, S. J.; Saghafi, A.; Williams, D. J. Greenhouse gas emissions from low-temperature oxidation and spontaneous combustion at open-cut coal mines in Australia. Int. J. Coal Geol. 2009, 78, 161–168.

(15) Kuenzer, C.; Stracher, G. B. Geomorphology of coal seam fires. Geomorphology 2012, 138, 209–222.

(16) Chen, G.; Ma, X.; Lin, M.; Peng, X.; Yu, Z. Pollutant emission characteristics and interaction during low-temperature oxidation of blended coal. J. Energy Inst. 2016, 89, 40–47.

(17) Liang, Y.; Liang, H.; Zha, S. Mercury emission from coal seam fire at Wuda, Inner Mongolia, China. Atmos. Environ. 2014, 83, 176–184.

(18) Day, S. J.; Carras, J. N.; Fry, R.; Williams, D. J. Greenhouse gas emissions from Australian open-cut coal mines: contribution from spontaneous combustion and low-temperature oxidation. Environ. Monit. Assess. 2010, 166, 529–541.

(19) Wang, Y. Research Progress and Prospect on Ecological Disturbance Monitoring in Mining Area. Acta Geodetica et Cartographica Sinica 2017, 46, 1705–1716.

(20) Stracher, G. B.; Taylor, T. P. Coal fires burning out of control around the world: thermodynamic recipe for environmental catastrophe. Int. J. Coal Geol. 2004, 59, 7–17.

(21) Liang, Y.; Zhang, J.; Wang, L.; Luo, H.; Ren, T. Forecasting spontaneous combustion of coal in underground coal mines by index gases: A review. J. Loss Prev. Process Ind. 2019, 57, 208–222.

(22) Yuan, L.; Smith, A. C. CFD modeling of spontaneous heating in a large-scale coal chamber. J. Loss Prev. Process Ind. 2009, 22, 426–433.

(23) Deng, J.; Xiao, Y.; Li, Q.; Lu, J.; Wen, H. Experimental studies of spontaneous combustion and anaerobic cooling of coal. Fuel 2015, 157, 261–269.

(24) Beamish, B. B.; Theiler, J. Coal spontaneous combustion: Examples of the self-heating incubation process. Int. J. Coal Geol. 2019, 215, 103297.

(25) Wu, Y.; Yu, X.; Hu, S.; Shao, H.; Liao, Q.; Fan, Y. Experimental study of the effects of stacking modes on the spontaneous combustion of coal gangue. Process Saf. Environ. Prot. 2019, 123, 39–47.

(26) Akgún, F.; Essenhügh, R. H. Self-ignition characteristics of coal stockpiles: theoretical prediction from a two-dimensional unsteady-state model. Fuel 2001, 80, 409–415.

(27) Xia, T.; Zhou, F.; Gao, F.; Kang, J.; Liu, J.; Wang, J. Simulation of coal self-heating processes in underground methane-rich coal seams. Int. J. Coal Geol. 2015, 141–142, 1–12.

(28) Zhuo, H.; Qin, B.; Qin, Q.; Su, Z. Modeling and simulation of coal spontaneous combustion in a gob of shallow buried coal seams. Process Saf. Environ. Prot. 2019, 131, 246–254.

(29) Miura, K. Simulation of Spontaneous Heating of a Small Fixed Bed of Dried Coal Exposed to a Flowing Wet Air Stream. Energy Fuels 2013, 27, 6148–6160.

(30) Jones, J. On the role of times to ignition in the thermal safety of transportation of bituminous coals. Fuel 2000, 79, 1561–1562.

(31) Li, L.; Jiang, D.; Beamish, B. Calculation of ignition times under adiabatic conditions by activation energy. J. China Coal Soc. 2010, 35, 802–805.

(32) Cerrien, K. The breeding fire control technology of coal seam; Coal Industry Press: Beijing, 1984.

(33) Yu, M.; Huang, Z.; Yue, C. Mathematic model for calculating the shortest coal spontaneous combustion time. J. China Coal Soc. 2001, 26, 516–519.

(34) Yang, Y.; Li, Z.; Hou, S.; Gu, F.; Gao, S.; Tang, Y. The shortest period of coal spontaneous combustion on the basis of oxidative heat release intensity. Int. J. Coal Sci. Technol. 2014, 24, 99–103.

(35) Yang, Y. J. China Univ. Min. Technol., 2009.

(36) Yang, Y.-l.; Li, Z.-h.; Pan, S.-k.; Gao, S.-y.; Wang, Y.-l. Oxidative heat release intensity in coal at low temperatures measured by the hot-wire method. Min. Sci. Technol. 2009, 19, 326–330.

(37) Yang, Y.; Li, Z.; Si, L.; Hou, S.; Li, Z.; Li, J. Study on test method of heat release intensity and thermophysical parameters of loose coal. Fuel 2018, 229, 34–43.

(38) Yang, W.; Luo, Y.; Vieira, B. Experimental technique and modeling for evaluating heat of rewetting effect on coals’ propensity of spontaneous combustion based on adiabatic oxidation method. Int. J. Coal Geol. 2018, 187, 1–10.

(39) Fabiańska, M.; Ciesielczyk, J.; Nądudvari, Á.; Misz-Kennan, M.; Kowalski, A.; Kruzewski, A. Environmental influence of gaseous emissions from self-heating coal waste dumps in Silesia, Poland. Environ. Geochim. Health 2019, 41, 575–601.

(40) Zheng, J.; Tian, K. Research on the relationship between industrial parameter and adsorption constant of coal. J. Henan Univ. Eng. 2017, 29, 20–23.

(41) Samper, J.; Yi, S.; Naves, A. Analysis of the parameter identifiability of the in situ diffusion and retention (DR) experiments. Phys. Chem. Earth 2010, 35, 207–216.