

Experimental study on interaction and kinetic characteristics during combustion of blended coals

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Abstract The combustion behaviors and kinetic parameters of three parent coals (A1, A2, and A3) and their blends (A1/A2 and A2/A3) have been evaluated under oxidizing atmosphere (O_2 and N_2 mixtures), using a non-isothermal thermo-gravimetric analyzer. The aim of this study is to investigate the interaction between the blended components during the process of co-combustion, and the effects of blending ratio and oxygen concentration (10, 15, and 21%) on combustion performance of blended coals. When high reactivity and low reactivity coals are co-combusted, double peaks are observed in the DTG curves, and significant interaction occurs in the temperature range between the two peaks (T_{p1} and T_{p2}). The activation energies obtained by Coats–Redfern method indicate that the activation energies of blended coals are lower than that of parent coals. The combustibility index S is used to evaluate the combustion performance of blended coals, and the results show the non-additive effects of the combustion characteristics of blended coals. The increased oxygen concentration results in a significant improvement of combustion performance of blended coals. In addition, as the blending ratio of high reactivity coal is increased, the oxygen can greatly enhance the combustion stability of blended coals.

Keywords Blended coals · Combustion · Kinetic analysis · Interactions

Introduction

In recent years, the utilization of blended coals is becoming common and attractive in pulverized fuel power stations [1–3]. It has been proved that coal blending is an effective way to improve combustion performance, meet pollutant emission limits, control ash deposition, extend the range of acceptable coals, and reduce the fuel cost [4]. Because coal-blend combustion is a very complex process, the underlying mechanisms are still poorly understood. Problems have been reported, such as high levels of unburned carbon in fly ash, flame instability, increase of slagging and fouling tendency, CO emissions, and plume opacity [5, 6]. At present, for most power plants the operation of coal-blend firing seems to be strongly dependent on the experience of individual power plant operators [1]. Thus, it is very essential to investigate the underlying mechanisms of coal-blend combustion.

In the last decades, extensive studies have been performed to characterize the combustion of blended coals. It has been recognized that the coal property parameters (e.g., proximate and ultimate analysis data, heating value, etc.) of blended coals remain additive, while many characteristics related to the combustion behavior exhibit non-additive [1, 5]. For example, ignition, flame stability, slagging and fouling, and pollutant emissions cannot be predicted by the additivity [3]. Especially when high and low reactivity coals are mixed, faster combustion of the high reactivity components might induce partial oxygen starvation of the low reactivity coals/chars [6], which causes a significant deviation between practical and predicted combustion performance.

Previously, several laboratory-scale devices have been used to study the combustion of blended coals: thermo-gravimetric analyzer (TGA), drop tube furnace (DTF)

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reactors, bomb-calorimeter-based test [3], and suspension firing reactors. Thermo-gravimetric (TG) analysis has been extensively applied to make a realistic assessment of the combustion of blends [7, 8]. TGA-based coal-blend combustion studies have proven that additive as well as synergistic effects [5]. In the case of binary blends, additivity of combustion behavior may be identified by the presence of two independent peaks in the DTG curves. Possible synergistic effects are indicated by the non-additivity of certain characteristics, such as ignition temperature and burnout time [6, 9]. DTF closely simulates the conditions prevailing in a pulverized fuel boiler, in particular the short residence time, high temperature and heating rate, and may be appropriate to study the combustion of coal blends, if enough interactions among the particles are ensured [10–12].

The interactions between the parent coals have been considered to be the main reason being responsible for the synergistic effects in coal-blend combustion. In this article, based on the TGA, the combustion process of blended coals is presented to analyze the interaction between parent coals. Combustion kinetic parameters are obtained by TGA to understand the combustion reactivity of blended coals. Studies on the effect of blending ratio and oxygen concentration on the interactions and combustion characteristics of blended coals have been reported in this article.

Experimental

Samples

Three coals A1, A2, and A3 widely used in Ningxia, China were selected as the parent coals in this study. Proximate and ultimate analyses along with net calorific values of the raw coals are shown in Table 1. The ratio of fixed carbon to volatile matter content (FC/VM) is referred to as fuel ratio, which can be used to evaluate combustion reactivity of fuels [13]. The smaller the fuel ratio is, the better the combustion reactivity of fuels is. The fuel ratios of three coals, A1, A2, and A3 are 1.79, 2.54, and 2.30, respectively. The samples of A1/A2 and A2/A3 have been blended in the proportion of 2:1, 1:1, and 1:2, respectively.

The raw coals were ground and sieved to obtain a particle size distribution of 100–125 μm . The coal mixtures were blended in desired proportions and manually homogenized.

TG analysis

NETZSCH-STA409C was used for TG analysis. In each experiment, 10 mg of sample coal was weighted precisely. Several different atmospheres ($\text{O}_2:\text{N}_2 = 10:90$, $\text{O}_2:\text{N}_2 = 15:85$, and $\text{O}_2:\text{N}_2 = 21:79$) was used as the carrier gas at a flow rate of 100 mL/min. The heating rate was set at 20 $^\circ\text{C}/\text{min}$ from the ambient temperature to 1200 $^\circ\text{C}$. Duplicate experiments for each test were performed to verify the reproducibility of the results. The mass loss (TG) and derivative (DTG) curves of the samples were represented as a function of temperature.

In order to find out the interactions between the components of the blends during the combustion process, the calculated DTG curves of the blends were obtained as the weighted sum of the DTG curves of each individual blended component [14]. Thus:

$$(dG/dt)_{\text{calc}} = x_A(dG/dt)_A + x_B(dG/dt)_B \quad (1)$$

$$x_A + x_B = 1, \quad (2)$$

where $(dG/dt)_A$ and $(dG/dt)_B$ are the mass loss rates of the individual coals, x_A and x_B are the proportions of the parent coals in the blends, respectively.

Determination of combustion characteristic temperature

As shown in Fig. 1, the combustion characteristic temperature was defined as follows: first, a vertical line passing through the differential thermo-gravimetric curve (DTG) peak point was made to meet the TG curve profile at point A, second, a tangent line from point A was made to meet the extended TG initial level line at point B, the corresponding temperature of the point B was defined as the ignition temperature (T_i); the corresponding temperature of the DTG peak point was defined as the temperature of the maximum combustion rate (T_p), finally, a tangent line of the tail end of the DTG defined was made to meet the extended DTG final level line at point C, the corresponding

Table 1 Proximate and ultimate analyses of coal samples

Samples	Proximate analysis/%				Ultimate analysis/%					$Q_{\text{net,ar}}/\text{MJ kg}^{-1}$
	FC_{ad}	VM_{ad}	A_{ad}	M_{ad}	C_{ad}	H_{ad}	O_{ad}	N_{ad}	S_{ad}	
A1	46.09	25.70	18.19	10.02	57.40	3.03	10.10	0.93	0.33	19.76
A2	45.97	18.12	35.16	0.75	54.94	3.10	4.61	1.09	0.35	19.76
A3	55.83	24.28	14.70	5.19	65.82	3.00	6.77	0.84	3.68	23.20

ad air dry basis, FC fixed carbon, VM volatile matter, A ash, M moisture, $Q_{\text{net,ar}}$ net calorific value

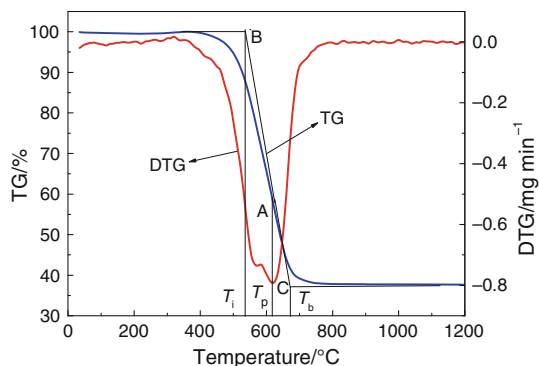


Fig. 1 Determination of combustion characteristic temperature

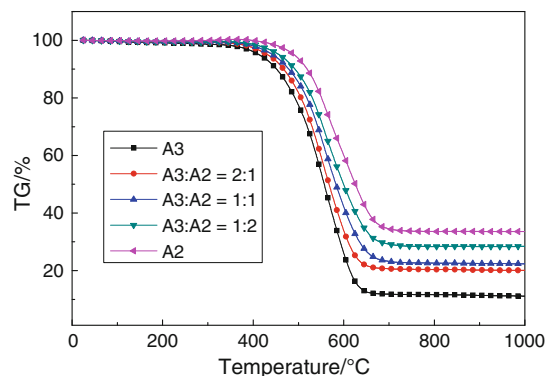


Fig. 4 TG curves of coal A2, coal A3, and their blends

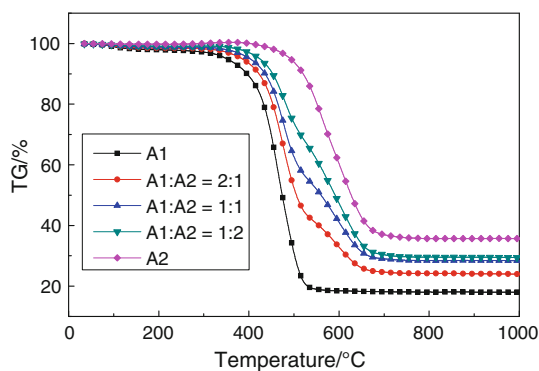


Fig. 2 TG curves of coal A1, coal A2, and their blends

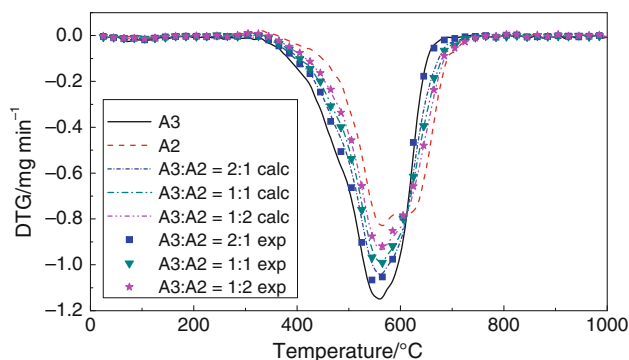


Fig. 5 DTG curves of coal A2, coal A3, and their blends

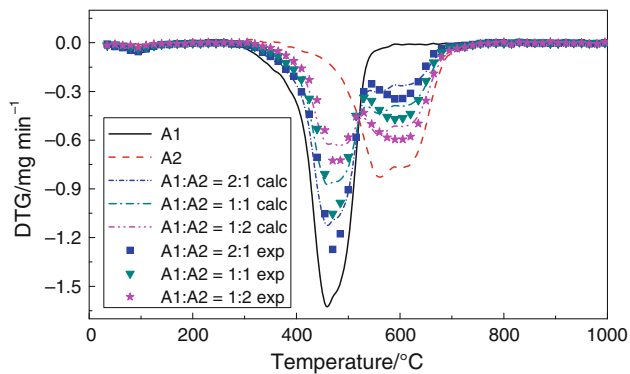


Fig. 3 DTG curves of coal A1, coal A2, and their blends

temperature of the point C was defined as the burnout temperature (T_b) [15].

Results and discussion

TG and DTG curves analysis

The TG and DTG curves of coal samples under the oxygen–nitrogen mixtures ($O_2:N_2 = 21:79$) are presented in Figs. 2, 3, 4, and 5. It is evident from the TG and DTG

curves that the mass loss process of coal samples can be divided into three stages: the stage of moisture evaporation and volatile decomposition, the stage of vigorous combustion of char, and the stage of burnout of char.

As can be seen from the TG curves in Fig. 2, the mass loss of coal A1 occurs obviously at low temperatures than coal A2 does, and the mass loss processes of the blended coals ($A1:A2 = 2:1$, $A1:A2 = 1:1$, and $A1:A2 = 1:2$) can be easily distinguished. As expected, the increased blending ratio of coal A2 causes a shift of the TG curves to a high temperature. This is mainly due to comparatively less volatile matter and more fixed carbon content in coal A2, which makes the combustion reactivity of coal A2 worse than that of coal A1.

Ignition temperature and burnout temperature are also used to describe the combustion performance of fuels. The low the ignition temperature is, the easier it is for fuels to ignite. The low the burnout temperature is, the less time for fuels to burn out, and the less combustible matter in the ash [16]. It is seen from Table 2 that the ignition temperature (T_i) and burnout temperature (T_b) of coal A1 are 418 and 514 °C, respectively, and those of coal A2 are 499 and 565 °C, respectively. It is also shown that the ignition and burnout performances of coal A1 are significantly better than those of coal A2. According to Table 2, it can

Table 2 Combustion characteristic temperatures of coal samples

Samples	$T_i/^\circ\text{C}$	$T_{p1}/^\circ\text{C}$	$T_{p2}/^\circ\text{C}$	$T_b/^\circ\text{C}$
A1	418	460	–	514
A1:A2 = 2:1	427	473	598	542
A1:A2 = 1:1	430	475	595	562
A1:A2 = 1:2	422	477	597	641
A2	499	565	–	664
A3	471	553	–	618
A3:A2 = 2:1	477	549	–	622
A3:A2 = 1:1	484	559	–	638
A3:A2 = 1:2	488	561	–	648
A2	499	565	–	664

be concluded that addition of high reactivity coals can improve combustion reactivity and shorten the burning time of blends. From Fig. 2 and Table 2, it is found that the ignition performance of blends is closer to that of high reactivity coal, and the burnout performance of blends is closer to that of low reactivity coal.

From the DTG curves in Fig. 3, a single peak is observed for the parent coals, and double peaks are shown for blended coals. The presence of two independent peaks indicates the additivity between the parent coals to a certain extent. The DTG data for blended coals shows a two-stage combustion process with peaks at approximately 475 and 595 °C, which may indicate that two different types of char reaction. This may be due to difference in indigenous properties of parent coals. It is also apparent that there is an overlapping of mass loss processes between the parent coals during the combustion of the blends, and this may cause interactions of each individual components. It is found from Table 2 that the first peak temperature (T_{p1}) of blends is closer to the peak temperature of high reactivity coal A1, and the second peak temperature (T_{p2}) is closer to the peak temperature of low reactivity coal A2, which suggests that the mass loss in the first stage should be mainly owing to coal A1, and the mass loss in the second stage should be owing to coal A2.

It is evident from Figs. 4 and 5, for coal A2, coal A3 and their blends that the mass loss processes are similar, and there is a single peak shown in the DTG profiles. The main cause is that the fuel ratios of two components are very close, 2.54 for coal A2 and 2.30 for coal A3. In Table 2, the differences both between the ignition temperatures and between the burnout temperatures of different blends are very small.

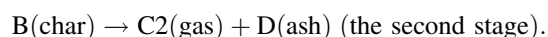
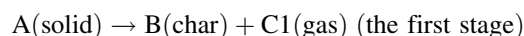
Interactions between the components of blends

It can be seen from the experimental and theoretical DTG curves of A1/A2 blends in Fig. 3 that in the first

temperature region, from ambient temperature to the first peak temperature (T_{p1}), there is no evident deviation between the experimental and calculated combustion rate; in the second temperature stage, from the first peak temperature to the second peak temperature (T_{p2}), the experimental combustion rate is obviously greater than the calculated combustion rate; in the last temperature region, no significant deviations are observed between the experimental and calculated DTG curves. This is mainly due to the absence of coal A2 combustion in the first temperature stage, the significant synergetic effects during the co-combustion process of the second temperature stage and the absence of coal A1 combustion in last temperature stage. According to the DTG results of A1/A2 blends, when high and low reactivity coals are co-fired, the significant interactions between the blend components will occur in the temperature stage from the first peak temperature (T_{p1}) to the second peak temperature (T_{p2}). The results are consistent with those obtained by other authors [10, 15], who observed important interactions between the blend components. From Fig. 5, no obvious deviations are observed between the experimental and calculated DTG curves of A2/A3 blends, which suggests that no significant interactions occur, and there exists the additive behavior of blend components with slight difference in reactivity during combustion process.

Analysis of reactivity and kinetic parameters

The coal combustion includes gas phase reactions between the volatiles released and oxygen at low temperatures, and the combustion of the char generated in the early stages of the solids degradation [17]. A two-stage reaction kinetics scheme consisting of two reactions is proposed for the thermal decomposition of the coal under an oxidative atmosphere [18, 19]. The two independent reactions are described as:



The approach used in this study to calculate the kinetic parameters was based on the Arrhenius equation, which has been used by other researchers [7, 14, 19–22] to obtain kinetic parameters of thermal events under combustion conditions. These two separate reactions are thought to be governed by the first-order Arrhenius law [23]. So the kinetics of the reaction is described as:

$$\frac{dx}{dt} = kf(x) \quad (3)$$

$$k = A \exp\left(-\frac{E}{RT}\right), \quad (4)$$

where $f(x)$ represents the hypothetical model of the reaction mechanism, k is the reaction rate, A is the pre-exponential

factor (s^{-1}), E is the activation energy (kJ mol^{-1}), R is the gas constant ($8.314 \text{ Jk}^{-1} \text{ mol}^{-1}$), T is the absolute temperature (K), t is the time (min), and x is the loss in mass fraction or mass conversion ratio, which can be calculated by the following relationship:

$$x = (m_0 - m_t)/(m_0 - m_f), \tag{5}$$

where m_0 is the initial mass of the sample, m_t is the mass of the sample at time t , and m_f is the final mass of the sample.

For a constant heating rate β (K min^{-1}) during combustion, $\beta = dT/dt$, Eq. 3 can be transformed into:

$$dx/f(x) = (k/\beta)dT. \tag{6}$$

Integrating Eq. 6 gives:

$$g(x) = \int_0^x dx/f(x) = A/\beta \int_{T_0}^T \exp\left(-\frac{E}{RT}\right)dT, \tag{7}$$

where $g(x)$ is the integral function of conversion.

Equation 7 is integrated by using the Coats–Redfern method [24], yielding:

$$\ln\left(\frac{g(x)}{T^2}\right) = \ln\left[\frac{AR}{\beta E}\left(1 - \frac{2RT}{E}\right)\right] - \frac{E}{RT}. \tag{8}$$

Since it can be demonstrated that for most values of E and the temperature range of combustion, the expression $\ln[AR/\beta E(1 - 2RT/E)]$ in Eq. 8 is essentially constant. If $\ln[g(x)/T^2]$ is plotted versus $1/T$, a straight line should be obtained. Moreover, if the correct $g(x)$ is used, the plot of $\ln[g(x)/T^2]$ against $1/T$ should give a straight line with a high correlation coefficient of linear regression analysis, from which the values of E and A can be derived. The activation energy E and pre-exponential factor A can be calculated from the slope and intercept of the line [25, 26].

In most dynamic studies using TGA, the chemical reaction model is the most frequently used [14, 27]. The chemical reaction model is described as:

$$f(x) = (1 - x)^n \tag{9}$$

where n is the order of reaction. Equation 9 can be transformed into:

$$\ln\left|\frac{-\ln(1 - x)}{T^2}\right| = \ln\left[\frac{AR}{\beta E}\left(1 - \frac{2RT}{E}\right)\right] - \frac{E}{RT} \quad (n = 1) \tag{10}$$

$$\ln\left|\frac{1 - (1 - x)^{(1-n)}}{T^2(1 - n)}\right| = \ln\left[\frac{AR}{\beta E}\left(1 - \frac{2RT}{E}\right)\right] - \frac{E}{RT} \quad (n \neq 1). \tag{11}$$

When different reaction orders were used to calculate A and E , results showed that the first-order reaction had the

best linear relationship, which indicated that the experiment was governed by the first-order Arrhenius law.

With the change of combustion temperature, the reaction mechanism might be different, so the kinetic parameters under different temperature stages should be analyzed separately [28, 29]. In this article, the combustion process of blended coals is divided into two temperature regions: the low temperature region (from T_i to T_p) and the high temperature region (from T_p to T_b). The kinetic parameters of coal samples in different temperature regions are shown in Table 3. It is apparent from Table 3 that the activation energies in the low temperature region are high than those in the high temperature region, which is in agreement with the study of other authors [15]. It indicates that in the low temperature region more energy is needed to maintain the combustion reaction; char combustion reaction mainly occurs in the high temperature region, however, it consumes a small amount of energy. The conclusions obtained here may be due to the high temperature condition and catalysis action of the mineral components in the high temperature region.

Since the activation energies in Table 3 are obtained in different combustion stages, they cannot be used to directly represent the reactivity of coal combustion during the whole combustion process. Therefore, in this article, the mean activation (E_m) proposed by Cumming [30] is introduced to evaluate the reactivity of the coals. E_m is described as:

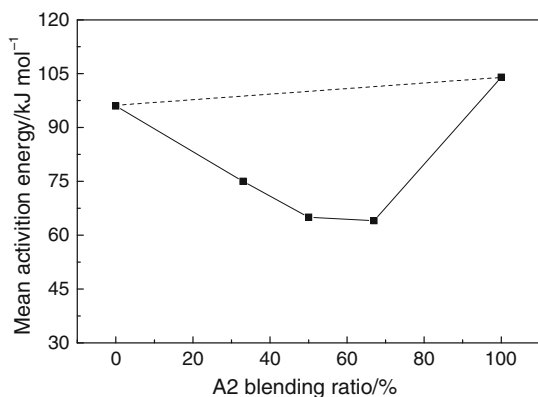
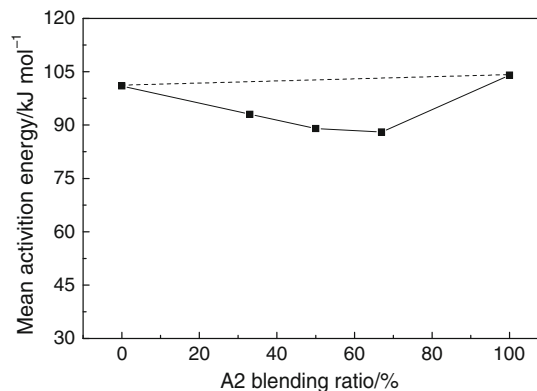
$$E_m = E_1F_1 + E_2F_2 + \dots + E_nF_n, \tag{12}$$

where $E_1 \sim E_n$ are the individual values of activation energy obtaining over each corresponding period of Arrhenius linearity, $F_1 \sim F_n$ are the mass fractions of combustible content of the samples burned during each region of Arrhenius linearity.

As shown in Figs. 6 and 7, dash lines represent the value of mean activation energy based on the weighted average; experimental values changing with blending ratio are also shown for comparison with the predicated values. Significant negative deviations are observed between the predicated and experimental mean activation energy of A1/A2 blends, which can be seen from Fig. 6. This might be attributed to the significant interactions between the high reactivity coal A1 and low reactivity coal A2. Since only weak interactions occur, the deviation between the predicated and experimental mean activation energy of A2/A3 blends is smaller than that of the A1/A2 blends, by comparing Figs. 6 and 7. It can be found that mean activation energy of blend coals does not always increase with the addition of low reactivity coal. Some researchers [31, 32] obtained similar results from their experiments, while others have reported that activation energy increases with the addition of low reactivity fuel [7, 8, 15]. It is

Table 3 Kinetic parameters of coal samples

Samples	Temperature region/°C	Mass fraction/%	Activation energy/E/kJ mol ⁻¹	Frequency factor ln(A)/s ⁻¹	Correlated coefficient/R
A1	418–460	12.21	102.19	15.7720	0.9813
	460–514	52.21	95.03	14.6060	0.9987
A1:A2 = 2:1	427–473	21.38	104.98	15.8590	0.9948
	473–542	25.31	51.01	6.4247	0.9928
A1:A2 = 1:1	430–475	17.18	103.58	15.0317	0.9992
	475–562	25.19	38.74	3.8256	0.9963
A1:A2 = 1:2	422–477	15.98	95.17	13.1895	0.9989
	477–641	41.94	52.66	5.9081	0.9906
A2	499–565	20.56	122.26	16.1340	0.9996
	565–664	33.30	93.36	11.7329	0.9993
A3	471–553	35.67	98.72	13.2721	0.9994
	553–618	24.80	105.19	14.2769	0.9999
A3:A2 = 2:1	477–549	22.59	101.23	14.4927	0.9962
	549–622	28.56	87.25	11.5486	0.9896
A3:A2 = 1:1	484–559	21.51	99.31	14.2199	0.9972
	559–638	31.73	82.65	9.2517	0.9896
A3:A2 = 1:2	488–561	23.45	97.26	13.4535	0.9984
	561–648	37.45	83.10	10.1064	0.9989
A2	499–565	20.56	122.26	16.1340	0.9996
	565–664	33.30	93.36	11.7329	0.9993

**Fig. 6** Mean activation energy of coal A1, coal A2, and their blends**Fig. 7** Mean activation energy of coal A2, coal A3, and their blends

noteworthy that the mean activation energy of the blended coals seems to be even less than the pure high reactivity coal, which may be attributed that the model of the chemical reaction mechanism of blended coals is different from that of the parent coal.

Analysis of combustion performance

The combustibility index S [33] is introduced as a criterion for evaluating fuel combustion performance, which is defined as:

$$S = \frac{R}{E} \frac{d}{dT} \left(\frac{dG}{dt} \right)_{T=T_i} \frac{\left(\frac{dG}{dt} \right)_{\max}}{\left(\frac{dG}{dt} \right)_{T=T_i}} \frac{\left(\frac{dG}{dt} \right)_{\text{mean}}}{T_b} = \frac{\left(\frac{dG}{dt} \right)_{\max} \left(\frac{dG}{dt} \right)_{\text{mean}}}{T_i^2 T_b}, \quad (13)$$

where $(dG/dt)_{\max}$ is the maximum combustion rate (mg min^{-1}), $(dG/dt)_{\text{mean}}$ is the average combustion rate, T_i is the ignition temperature ($^{\circ}\text{C}$), and T_b is burnout temperature ($^{\circ}\text{C}$). This index S , which encompasses the ignition temperature, the combustion rate and burnout temperature, is a comprehensive parameter, used here to

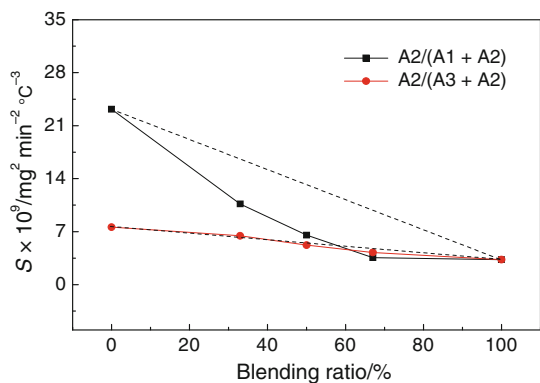


Fig. 8 Combustibility index of blended coals

compare the combustion performance of blended coals. The greater the value of the index S is, the high the combustion reactivity is.

Effect of blending ratio

Blending ratio plays a crucially important role in combustion performance of blended coals. If the blending ratio is chosen unreasonably, the combustion condition would become worse. Figure 8 gives the effects of blending ratio on combustion performance. It is evident that the combustion performance shows a downtrend with increased proportion of low reactivity coal, and is closer to that of low reactivity coal, which is mainly attributed to less volatile matter and more fixed carbon content in low reactivity coal. The results are consistent with the reports by Xu et al. [15]. It could be concluded that the addition of high reactivity coals can improve the combustion performance of blended coals. The dash lines in Fig. 8 show the value of combustibility index based on the weighted average. It is obvious that the combustion performance of A1/A2 blends displays non-additive behavior induced by synergistic effects, and that of A2/A3 blends shows approximately additive behavior. These could be verified by the results obtained from the mean activation energy of coal samples.

Effect of oxygen concentration

In the furnace of power plant boiler, the combustion process of the pulverized coal almost occurs in the low oxygen concentration environment (3–21%). Consequently, it is needed to investigate the combustion performance of blended coals in the low oxygen concentration environment. The combustion of coal A1, coal A2, and their blends under different oxygen concentration (10, 15, and 21%) have been carried out in the TGA. Figure 9 presents the combustion performance of blended coals under different oxygen concentrations.

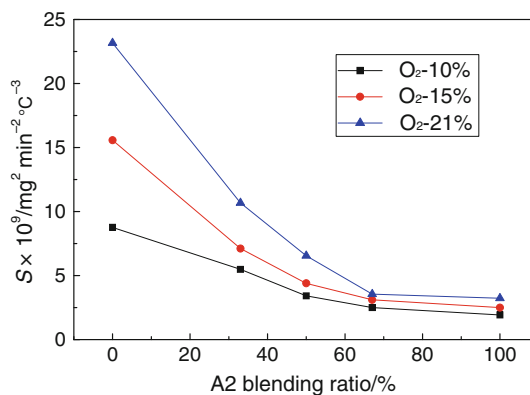


Fig. 9 Combustibility index of coal A1, coal A2, and their blends

It can be observed from Fig. 9 that the combustion performance of blended coals worsens with the decreased oxygen concentration under the same blending ratio, which can be explained like that the oxygen insufficiency makes the ignition and burnout difficult. It is also apparent that the influence of oxygen concentration on the combustion performance of blended coals is weakened as the blending ratio of high reactivity coal decreases. When high and low reactivity coals are co-fired, the faster combustion of high reactivity components may induce oxygen insufficiency of low reactivity chars in the later period of combustion, which makes the burnout performance worse. Consequently, with decreased blending ratio of high reactivity coals, the competition for oxygen with low reactivity components decreases. It can be concluded that high oxygen concentration atmosphere can significantly improve the combustion stability of blended coals, especially as the blending ratio of high reactivity coal is increased.

Conclusions

The combustion behaviors of the parent coals (A1, A2, and A3) and their blends (A1/A2 and A2/A3) have been studied using a TGA. The objectives of the study are to investigate the interactions between the blended components, the effects of the blending ratio on combustion performance and kinetic characteristics, and the effects of oxygen concentration on combustion performance.

Addition of high reactivity coals can improve combustion reactivity and shorten the burning time of blended coals. The ignition performance of coal blends is closer to that of high reactivity coal, and the burnout performance of blends is closer to that of low reactivity coal. When high and low reactivity coals are co-fired, the obvious double peaks are observed in the DTG curves, and the significant interactions occur in the temperature stage from the first peak temperature (T_{p1}) to the second peak temperature

(T_{p2}). Addition of high reactivity coals can decrease the activation energy required in the combustion reaction.

The combustion performance shows non-additive behavior induced by synergistic effects when high and low reactivity coals are mixed, while additive behavior remains when the similar reactivity coals are mixed. Besides, the mean activation energies of blended coals are lower than that of parent coals. With the increase of oxygen concentration, the combustion performance of blended coals is improved significantly. In addition, as the blending ratio of high reactivity coal is increased, the oxygen can greatly enhance the combustion stability of blended coals.

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