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Study on Combustion Characteristics and Kinetics Analysis of Zhundong Lignite

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Abstract. The high moisture and low calorific value of Zhundong lignite restricts its long-distance transportation and large-scale application. In this paper, the elemental composition changes and combustion characteristics of the upgraded lignite were studied, and then the combustion reaction kinetics of the upgraded lignite was analyzed by the Flynn-Wall-Ozawa (FWO) method. The results show that with the increase of the upgrading temperature, the content of volatiles and water in the coal decrease, which leads to an increase in the proportion of carbon, nitrogen and sulfur, and a decrease in the content of hydrogen and oxygen. The thermogravimetric curve of the upgraded lignite increases between 573K and 673K as the heating rate increases due to the influence of thermal buoyancy and inhalation gain. The burnout index H_j obviously increases with the increase of the heating rate, and the burnout condition of the coal is obviously improved. The apparent activation energy obtained at the same conversion rate and decreases with the increase of the upgrading temperature, which proves that the increase of the upgrading temperature reduces the apparent activation energy of the upgraded lignite.

1. Introduction

Fossil fuel will still play an important role in the future power generation industry [1]. Zhundong lignite is a kind of lignite with high moisture content, low calorific value and high alkali metal content, which is produced in Xinjiang, China [2]. Due to the inconvenience of transportation, low quality, slagging and other problems, Zhundong lignite is greatly restricted in the utilization process [3]. However, the low price of lignite makes it have unique advantages in the energy market. Therefore, how to make full use of lignite has important economic and social value.

Upgrading technology is a common pretreatment technology of lignite. Pre-drying and quality improvement technology include evaporation drying technology [4] and non-evaporation drying technology. Methods of evaporation dehydration include mixing grinding and drying, fluidized bed drying, pressurized steam drying, microwave drying and solar drying [6]. Non-evaporation drying methods include Reynard process, K-fuel process, hydrothermal dehydration, lignite technology upgrading, mechanical hot extrusion, etc. [6]. Among them, the technology of pre-drying and upgrading lignite with flue gas contain more waste heat produced by combustion in power plant (i.e. the waste gas



that we usually think cannot be used) has a good application prospect. Zhu Xin et al. [7] studied the application and thermodynamic performance of pre-drying technology in wood fuel power plant. Xiaoqu Han et al. [8] have studied the recovery and kinetic analysis of water after pre-drying lignite, but have little knowledge of the change of combustion properties of coal during pre-drying. Hongyu Zhao [9] et al. Studied the drying and dynamic characteristics of lignite in different atmospheres, but did not explore the change of combustion characteristics of lignite in the presence of different oxygen or carbon dioxide. Thermogravimetric analysis has been widely used in the kinetic analysis process (such as co-pyrolysis of biomass and coal in different atmospheres [10], CO pyrolysis of coal and semi coke [11]). Most of the work on the thermogravimetry of pre-dried coal is focused on the change of physical and chemical properties of lignite samples before and after pre-drying. However, there is very little information about the influence of different upgrading conditions on the combustion characteristics of lignite. Combined with the theory of thermal analysis dynamics, the combustion characteristics and combustion dynamics parameters of the samples are obtained by analyzing the combustion thermogravimetry curve, so as to evaluate the combustion properties of the improved lignite.

In this paper, Zhundong lignite was improved by simulated flue gas of different temperatures, and the thermogravimetric combustion experiment was carried out by synchronous thermal analyzer, and the combustion kinetics was analyzed by FWO method.

2. Experiment

2.1. Quality improvement system and samples

The research object of this paper is Zhundong lignite from Xinjiang. Use crusher to crush Zhundong block raw coal, and then use vibrating screen machine to screen it into 0.075-0.125mm particle size for standby. See Table 1 for industrial analysis and element analysis results of coal sample.

Table 1. The proximate and ultimate analysis of ZD lignite

Proximate analysis (%)				Ultimate analysis (%)			
M_{ad}	V_{ad}	A_{ad}	FC_{ad}	C_d	H_d	O_d	N_d
6.33	51.64	29.51	12.52	75.64	4.09	14.00	0.79

The screened Zhundong raw coal is improved by using the experimental system shown in Figure 1. Take $3g \pm 0.1mg$ of raw coal and put it into the quartz reactor, and inject 300ml / min nitrogen into the quartz reactor for evacuation. The crucible resistance furnace was heated to 423K, 623K and 773K respectively, and the constant temperature was 30min. Adjust the mass flow meter, the air flow into the quartz reactor is 300ml / min, N_2 (95%) / O_2 (5%) atmosphere (marked as II atmosphere in this experiment). Then put the quartz reactor into the furnace for 30min, take out the quartz reactor, and put the sample into the plastic bag for standby after the nitrogen of 300ml / min is introduced to cool the sample to room temperature. The samples were labelled as II-423K, II-623K and II-773K respectively.

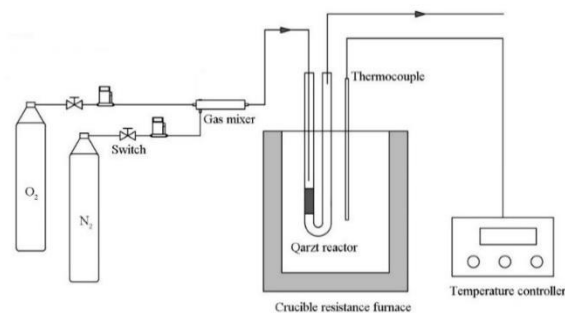


Figure 1. The lignite upgrade experiment rig

2.2. Thermogravimetric experiment

The combustion characteristics of the improved lignite under different conditions were analysed by the synchronous thermal analyzer. The non-isothermal thermogravimetry is to make the sample combust under the condition of linear temperature rising. The mass of the sample decreases with the increase of temperature. The combustion reaction characteristics of the sample are characterized by the change of conversion rate. The experimental equipment is STA449F5 type atmospheric synchronous thermal analyzer produced by NETZSCH.

Preheat the Al_2O_3 crucible to 1000K, then make it cool to room temperature and take out the crucible. Weigh $10 \pm 0.005\text{mg}$ of sample put in the crucible with air flow of 100ml / min, set heating temperature range of 30 °C ~ 1000 °C, and heating rate of 10K / min, 20K / min and 40K / min respectively.

The sample is marked on the basis of the rising temperature rate. For example, the 10K / min sample of II-423K is marked with II-423K-10.

2.3. Data processing

The ignition temperature reflects the initial temperature of combustion reaction. In this paper, TG-DTG tangent method is used to determine the ignition temperature. The TG and DTG curves of coal combustion reaction are obtained from the thermogravimetry experiment. The weight loss peak point of the DTG curve is used as the vertical line, the intersection point of the vertical line and the TG curve is used as the tangent line of the TG curve, and the temperature corresponding to the intersection point of the tangent line and the weight loss starting parallel line is the ignition temperature T_i .

The burnout index [12] H_j is the characteristic parameter to judge the burnout performance of pulverized coal. The formula of H_j is

$$H_j = \frac{(\frac{dw}{dt})_{\max}}{T_i \cdot T_{\max} \frac{\Delta T_h}{\Delta T}} \quad (1)$$

In this paper, multiple scanning rate method is used, that is, TGA curves obtained at different heating rates are used to analyze combustion dynamics. Because the data at the same conversion rate on multiple TGA curves are usually used in calculation, it is also called equal conversion method [13]. The outstanding advantage of using the equal conversion method to estimate the activation energy is that the activation energy value can be calculated directly without considering the reaction mechanism function, so the error caused by the wrong choice of the reaction mechanism function can be effectively avoided.

The reaction rate of combustion, defined as the change of unit time, is calculated according to the following formula:

$$\frac{d\alpha}{dt} = k(T)f(\alpha) \quad (2)$$

$$\alpha = \frac{m_i - m_a}{m_i - m_f} \times 100\% \quad (3)$$

Where α is the conversion rate, t is the time, $T(K)$ is the absolute temperature, and $k(T)$ is the rate constant about the temperature. m_0 , m_a and m_f are the initial mass, the current mass and the residual mass after upgrading, respectively.

According to Arrhenius equation, the expression of $k(T)$ is

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

Where A is the frequency factor (1 / min), E is the activation energy, in kJ / mol, R is the molar gas constant, and the value is $8.314 \text{ J/mol} \cdot \text{K}^{-1}$.

The combustion activation energy of the samples was calculated by Flynn Wall Ozawa (FWO) method [14]. The equation of Flynn wall Ozawa (FWO) method can be expressed as follows:

$$\ln(\beta) = \ln\left[\frac{0.0084AE_\alpha}{RG(\alpha)}\right] - 1.5016 \frac{E_\alpha}{RT} \quad (5)$$

At the same conversion rate α , $G(\alpha)$ is a fixed value. From the equation, it can be seen that $\ln(\beta)$ has a linear relationship with $1/T$. The apparent activation energy value E_a under the conversion rate α can be calculated by the slope of the straight line.

3. Results and discussion

3.1. Element analysis

The elemental analysis of different lignite is carried out by using the elemental vario macro cube constant element analyzer, and the data is listed in Table 2. It can be seen from the data in the table that with the increase of upgrading temperature, the contents of carbon, nitrogen and sulfur elements increase, while the contents of hydrogen and oxygen elements decrease. This is because with the increase of the temperature of upgrading, the amount of volatile matter and water in the coal decrease. At this time, due to the low oxygen content, the amount of carbon combustion loss is relatively small, which leads to the increase of the proportion of carbon, nitrogen and sulfur. Because the change of the content of elements in the improved lignite samples will have an important impact on its combustion properties, especially the increase of the proportion of combustible elements such as carbon will increase the calorific value of the fuel, and the proportion of different elements will also affect the combustion difficulty of the improved lignite.

Table 2. Ultimate analysis of upgraded lignite

	II-423K	II-623K	II-773K
C _d (%)	58.06	59.43	64.35
H _d (%)	4.016	3.162	2.12
S _d (%)	0.88	1.01	1.03
N _d (%)	0.194	0.242	0.319
O _d (%)	36.85	36.156	32.181

3.2. Thermogravimetric Characteristics

3.2.1. TG and DTG curves of lignite combustion at 10 K / min

Figure2 shows the TG and DTG curves of the improved lignite during combustion at different temperatures, in which the solid line is the TG curve and the dotted line is the DTG curve. It can be seen from Figure2 that the thermogravimetry curve of lignite with different upgrading temperatures has little difference at the initial stage of reaction, but it is arranged from low to high in the order of 437K, 623K to 773K from 573K to 673K. This is due to the different rate of combustion and heat release of the samples, resulting in the different magnitude of the heated buoyancy. The samples with high heating buoyancy are the samples with 773K upgrading temperature, and the TG curve of the samples has a large upward shift. At the same time, in the process of upgrading lignite, water and volatile matter will be lost, resulting in a relatively developed pore structure. In the process of combustion, a small amount of oxygen absorption or nitrogen absorption will increase weight, which will also cause the TG curve to shift upward [15]. With the increase of the temperature of upgrading, the moisture and a large amount of volatile matter released from lignite will increase, the coke pore will be more developed and more gas will be absorbed. The thermogravimetric curve shows that the maximum weight loss rate is reached between 673K and 773K, indicating that the combustion reaction is the most intense at this time. The coincidence degree of TG and DTG curves after 773K is high, which shows that the combustion process has little difference.

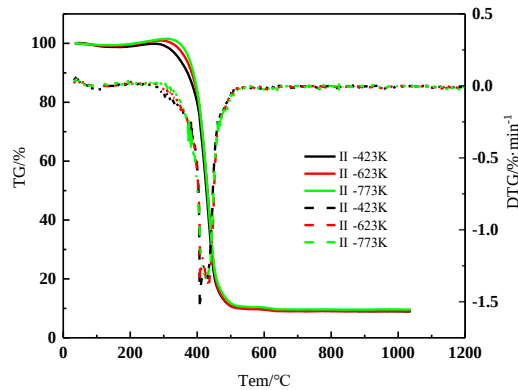


Figure 2. TG and DTG curves of upgraded ZD lignite under different temperature

3.2.2. TG and DTG curves of the same lignite under different heating rates

As shown in Figure 3, the slope of TG curve decreases with the increase of heating rate between 673K and 873K, and the peak point of DTG curve increases with the increase of heating rate. This is because with the increase of the heating rate, the temperature difference for inside and outside of the sample particles increases, resulting in the thermal lag phenomenon. The internal temperature of the particles is relatively low, and the reaction rate is relatively smaller than outside, resulting in the delay of volatilization analysis, and the temperature corresponding to the maximum weight loss rate point increases [16]. It can be seen clearly in the figure that the reaction starting temperature of the sample with lower heating rate is lower, because the sample has relatively sufficient time to absorb oxygen and be heat up under the condition, and then starts the combustion reaction. The peak area of DTG curve increases with the increase of heating rate. The peak area of DTG curve corresponds to the mass loss of sample during combustion, so it means that when the weight loss is reached during the reaction, the sample with higher heating rate will have lower temperature.

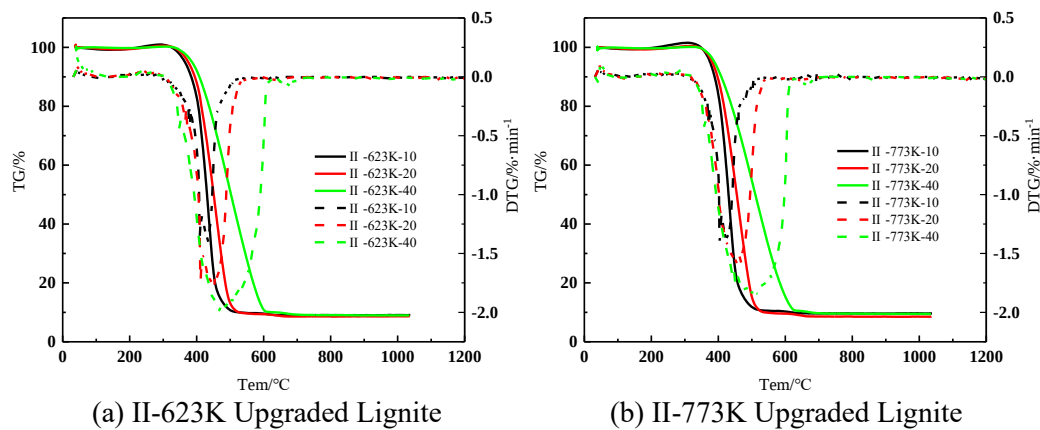


Figure 3. TG and DTG curves of upgraded ZD lignite at different heating rates

3.3. Combustion characteristic parameters

The combustion characteristic parameters of raw coal and improved lignite with different heating rates are shown in Table 3. The combustion characteristics of the same sample with different heating rates are obviously different. The maximum weight loss rate temperature T_{max} increases with the increase of heating rate, which is also due to the effect of thermal lag on sample combustion. The burnout index HJ increases obviously with the increase of heating rate, indicating that the burnout of coal increases obviously with the increase of heating rate. But the Δt of lignite increased from 423K, 623K to 773K at different temperatures. This is because the higher the temperature of lignite is, the more complete the non-combustible material such as water is, the more the proportion of the remaining coal combustible

material is. This is also consistent with the results of element analysis that the higher the temperature of lignite is, the higher the content of carbon, nitrogen and sulfur is, and the lower the content of hydrogen and oxygen is.

Table 3. Combustion characteristic parameter of upgraded lignite

Sample No.	T_i	T_{max}	ΔT	$H_j(\times 10^{-6})$
RC-10	384.4	405.9	36.6	0.90
RC-20	397.4	444.4	77	1.53
RC-40	385.2	455	180	3.43
II-423K-10	382.5	407.9	41.9	0.96
II-423K-20	391.2	437.4	89	2.01
II-423K-40	389.9	459.9	180	3.47
II-623K-10	394.7	430.3	49.4	0.90
II-623K-20	395.7	453.1	86	1.77
II-623K-40	394.9	466.3	195	3.60
II-773K-10	394.2	429.9	50	0.85
II-773K-20	397.2	456.9	100	1.55
II-773K-40	400.6	499.4	215	3.22

3.4. Kinetic parameters

The FWO method is used to fit the combustion activation energy of raw coal and improved lignite samples, and the fitting image is shown in Figure 4.

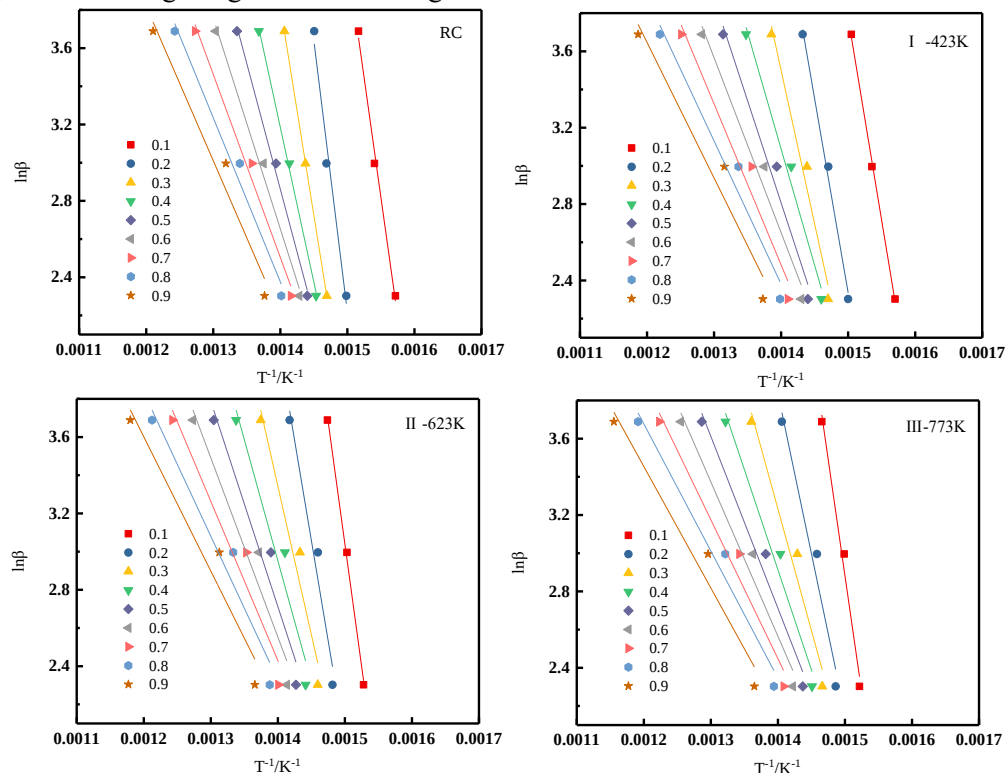


Figure 4. Fitting images of sample combustion activation energy by FWO method

According to the data fitting in Figure4, the activation energy value is calculated by FWO method, which is arranged in Table 4.

As shown in Figure 4, the fitting effect is better at the lower conversion rate, while the straight line fitted at the higher conversion rate deviates from the data greatly.

Except that the value of $\alpha = 0.1$ of RC sample is smaller than that of $\alpha = 0.2$, the activation energy of other samples decreases from 0.1 to 0.9. With the increase of conversion, the activation energy of the sample decreased obviously. The higher the apparent activation energy is, the lower the activation energy is and the easier the combustion reaction is.

The apparent activation energy values of the samples with the same conversion rate at different upgrading temperatures decrease with the increase of the upgrading temperature, which proves that the increase of the upgrading temperature reduces the apparent activation energy of the improved lignite and enhances the reactivity of the improved lignite.

Table 4. The activation energies obtained from FWO method for pyrolysis of biomass samples

α	RC	II-423K	II-623K	II-773K
0.1	197.82	237.28	202.96	255.43
0.2	224.78	190.25	165.67	161.16
0.3	173.24	140.46	123.04	113.8
0.4	128.16	113.7	100.23	93.58
0.5	104.04	94.2	84.96	77.27
0.6	87.32	81.53	74.15	68.31
0.7	75.77	71.57	65.88	61.26
0.8	67.9	64.61	59.56	55.33
0.9	63.92	60.56	55.57	51.95

4. Conclusion

With the increase of upgrading temperature, the amount of volatile matter and water in the coal decrease, so the proportion of carbon, nitrogen and sulfur increases, and the content of hydrogen and oxygen decrease.

The thermogravimetric curve of the improved lignite increases with the increase of heating rate due to the influence of thermal buoyancy and suction between 573K and 673K. The slope of TG curve decreases with the increase of heating rate between 673K and 873K, and the peak point of DTG curve increases with the increase of heating rate.

The maximum weight loss rate temperature $T_{\max}$ and the burnout index H_j increases with the increase of heating rate. Indicating that the burnout of coal increases with the increase of heating rate.

The value of apparent activation energy of lignite samples with the same conversion rate under different upgrading temperature decreases with the increase of upgrading temperature, which proves that the increase of upgrading temperature reduces the apparent activation energy of lignite and enhances the reactivity of lignite.

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