

THERMAL ANALYSES OF THE LIGNITE COMBUSTION IN OXYGEN-ENRICHED ATMOSPHERE

by

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Original scientific paper
DOI: 10.2298/TSCI141005007C

In this work, the oxygen-enriched combustion behavior of indigenous lignite was measured by a thermogravimetric analyzer. Combustion tests were carried out in N₂/O₂ atmosphere with an oxygen content ranging from 21 to 70 vol.%. The data show that the calorific value per unit time of the lignite was improved with increased oxygen concentration. In order to reveal the oxygen-enriched combustion process in detail, a new index of characteristic temperature T_c was defined to indicate the fixed carbon behavior during burning, and the combustibility indexes D' , R_w , and S were calculated to determine the combustion performance. The functional relation between calorific value per unit time and oxygen concentration was determined by non-linear fitting method. The results indicated that the relationship between calorific value per unit time and oxygen concentration fit the exponential function exactly.

Key words: *thermogravimetric analyzer, lignite, oxygen-enriched, calorific value per unit time*

Introduction

Coal is expected to increase in importance and become the predominant fossil fuel in use for at least the next 50 years [1]. The lignite coal regions of Inner Mongolia account for 75% of the total lignite quantity in China [2], which will need to be utilized in several ways. Currently it is mostly burned for power generation. However, its low heating value limits its wide utilization in combustion [3], such as in calcining cement clinker.

Thermal analysis of solid materials is very important for understanding their thermal behavior. TG/DTG/DSC analyses of coals have been carried out under different experimental conditions by any researches [2, 4-6]. Combustion characteristics such as heat content, ignition, and burn-off temperatures and the maximum rate of mass loss are important in coal utilization. These features can be determined from the thermogravimetric analyzer (TG) and DTG curves of the coal samples [7].

Oxygen-enriched technology is widely used in combustion system on experimental-scale [3, 8-11] to improve the burning rate and to reduce the emission of pollutants. Selcuket and Yuzbasi [12] performed oxygen-enriched combustion experiments using Turkish lignite in an

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O₂/N₂ environment at an oxygen concentration of 30 vol.%. Their results show that as the O₂ concentration increases, thermogravimetric curves shift through a lower temperature zone, the peak and burnout temperatures decrease, the mass loss rate increases, and complete combustion is achieved at low temperatures over a short time. Zhou *et al.* [13] conducted experiments on two types of lignite in an O₂/CO₂ environment using the TG method; the oxygen concentration ranged from 31 vol.% to 100 vol.%. They revealed that the coal combustion process is strongly affected by the O₂ concentration, and a lower O₂ concentration had an obvious effect on the TG curves. Niu *et al.* [14] studied three types of pulverized coal in an oxy-fuel atmosphere by thermogravimetric analysis and suggested that the O₂ concentration should be less than 40 vol.% in oxy-fuel combustion. Li *et al.* [15] performed TG analysis with one coal type and four different O₂ concentrations. Those results showed that when the O₂ concentration increased to a maximum of 80 vol.%, the combustion rate increases and the burnout time decreases. Daood *et al.* [16] investigated the effect of 28%, 30%, and 35% overall oxygen concentrations in an 20-kW down-fired pilot-scale combustion test facility, and oxygen was also provided at low stoichiometric ratios. The previous studies focus on bituminous coal and anthracite coal combustion or used an O₂/CO₂ environment for combustion. Few studies have reported lignite combustion in O₂/N₂ conditions or given the combustion laws in an oxygen-enriched environment. O₂/N₂ condition cuts nitrogen oxide emissions.

The main purpose of this work is to investigate the combustion behavior of lignite in an oxygen-enriched atmosphere, choose the appropriate O₂ concentration using the calorific value per unit time (CVT) value, explore the oxygen-enriched combustion laws of lignite with no-linear fitting method, and provide data for the performance of lignite combustion in calcining clinker.

Experimental

Materials

Three types of typical indigenous lignite were obtained from the Pingzhuang underground coal mine (PZ), Huolinhe opencast mine (HLH), and Zhalainuoer opencast mine (ZLNE) in Inner Mongolia in China. The coal samples were first crushed and pulverized with a 5EPC1×100 pulverizer, then sieved to less than 80 μm. Proximate analysis of the lignite was performed by a 5E-CHN2000 instrument, and ultimate analysis was carried out with a 5E-MAG6700 element analyzer and 5E-IRS3000 sulfur analyzer. A 5E-KCIV calorimeter was

Table 1. Proximate and ultimate analysis of lignite samples

Proximate [%]					$Q_{\text{net,ar}}$ [MJkg ⁻¹]
Sample	M_{ad}	VM_{d}	FC_{d}	A_{d}	
PZ	6.28	32.73	45.39	21.89	17.263
HLH	3.15	34.39	37.36	28.25	12.529
ZLNE	4.90	39.92	48.29	11.78	16.834
Ultimate [%]					
Sample	C_{d}	H_{d}	O_{d}	N_{d}	S_{d}
PZ	55.67	3.54	17.77	0.66	0.48
HLH	51.72	3.70	15.27	0.68	0.41
ZLNE	63.07	3.88	20.52	0.60	0.14

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

utilized to determine the calorific values. Proximate analyses data, ultimate analyses data and calorific value are briefly summarized in tab. 1. As seen in the table, lignite can be characterized by its low calorific value. The low calorific value can not meet the requirement in calcining clinker.

Methods

In this paper, lignite combustion and pyrolysis TG/DTG/DSC analyses were performed on NETZCH-STA409PC instruments. Approximately 10 mg of a coal sample was heated from room temperature to 1273 K during each experiment, with a heating rate of 10 Kmin⁻¹. Pyrolysis tests were carried out under a N₂ atmosphere, which is the diluting gas in the experiment. Combustion tests were conducted in an O₂/N₂ atmosphere with O₂ concentrations of 21%, 30%, 40%, 50%, 60%, and 70 vol.%. The total gas flow was set to 80 mLmin⁻¹ in both pyrolysis and combustion tests.

The TG/DTG/DSC profiles obtained during pyrolysis and combustion experiments were used to determine characteristic parameters such as initial decomposition temperature (T_i), peak temperature (T_{max}), ignition temperature (T_{ig}), and burnout temperature (T_b) of the fuel samples. T_i represents the initiation of mass loss devolatilization and is defined as the temperature where the TG curve of pyrolysis diverged from combustion curve after the moisture loss point. T_{max} is the point where the maximum reaction rate occurs. T_{ig} is defined as the temperature at which the coal starts to burn. It is taken as the position immediately after the combustion reaction beginning when the rate of mass loss is 0.1% K⁻¹. The last characteristic temperature T_b is the burn out temperature, which represents the point at which sample oxidation is completed. It is taken as the point immediately before the reaction ends when the rate of mass loss is 0.1% K⁻¹ [17]. The characteristic temperatures have been described in a number of other investigations [18-21]. There are no parameters to characteristic the combustion process from ignite point to T_{max} point. A new characteristic temperature point was defined through DSC curve. All experiments were repeated, and the mean values were used provided that the deviations were within 5%.

The point T_c is the starting point of the second exothermic peak in DSC curve, which is the temperature at which the carbon starts burning. Each point is illustrated in fig. 1.

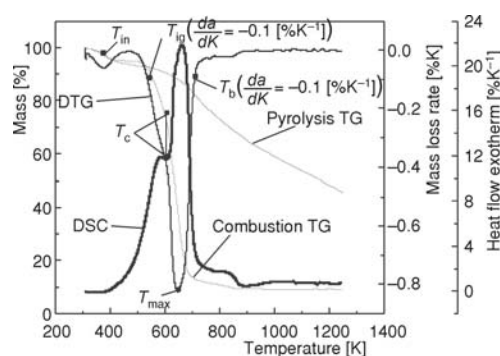


Figure 1. The points representing the characteristic temperatures

Results and discussion

Thermal tests of pyrolysis process of the lignite in N₂ environment

Pyrolysis is the initial process of coal combustion, which plays a crucial role in determining flame stability, ignition, and product distribution [22]. The TG, DTG, and DSC profiles of pyrolysis tests are shown in figs. 2, 3, and 4, respectively.

As shown in fig. 2, the mass of the residual solid was different for the three samples, which was consistent with their volatile content shown in tab. 1. From DTG profiles and DSC

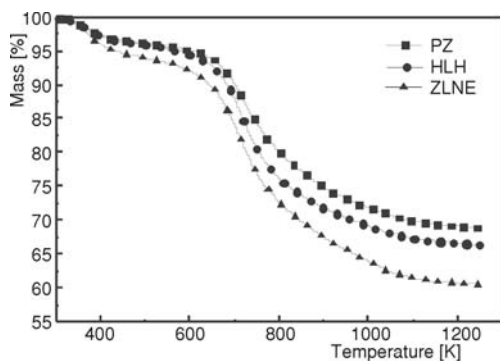


Figure 2. TG profiles of the lignite pyrolysis process

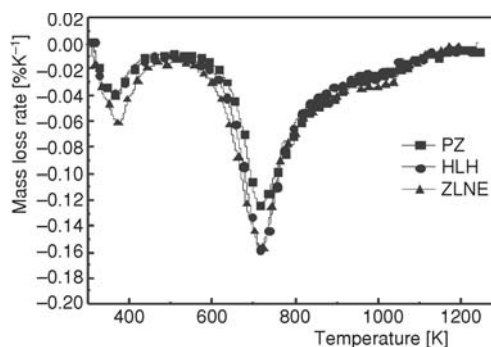


Figure 3. DTG profiles of the lignite pyrolysis process

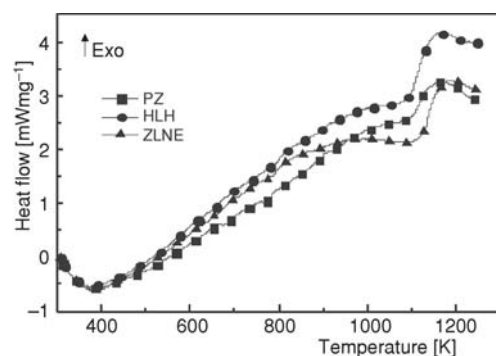


Figure 4. DSC profiles of the lignite pyrolysis process

decomposition process of lignite is only affected by the temperature and is not related to the volatile content.

Thermal tests of oxygen-enriched lignite combustion process

Thermogravimetric experiments were conducted to determine the oxygen-enriched combustion behavior of lignite. The TG, DTG, and DSC curves of lignite samples under different gas environments are presented in figs. 5, 6, and 7. From the curves the combustion process can be divided into three stages. The first stage is moisture evaporation and a small part volatile decomposition. The second stage is the volatile decomposition and char combustion. The last stage is burn out.

In fig. 5, the curves overlap each other at different oxygen concentrations in the first stage, which indicated that the mass loss of moisture and initial reaction of volatile compound decomposition were not affected by the oxygen concentration. Comparing the pyrolysis TG curve and combustion TG curve, the pyrolysis curve superposed with combustion curves in the moisture evaporation process. Immediately after the moisture evaporation process, the mass loss continued in the pyrolysis curves. However, the slight mass increased in the same temperature zone in combustion curves. This phenomenon might be attributed to the activity groups on the surface of the coal combination with the oxygen, which would offset the mass loss of devolatilization by the oxygen molecule.

profiles in figs. 3 and 4, it was found that the initial reaction of pyrolysis is an endothermic reaction at about 380 K, there appeared a weight loss peak in the DTG profiles either. The phenomena can be attributed to moisture loss and release the gas adsorption in the coal. The second weight loss peak in the DTG profiles is the stage of vigorous reaction. Figure 3 indicates that the three samples had the same values of T_i and T_{max} . Higher volatile content results in higher rate of mass loss before T_{max} , however it is generally consistent above the T_{max} of three samples. The results indicate that the decomposition

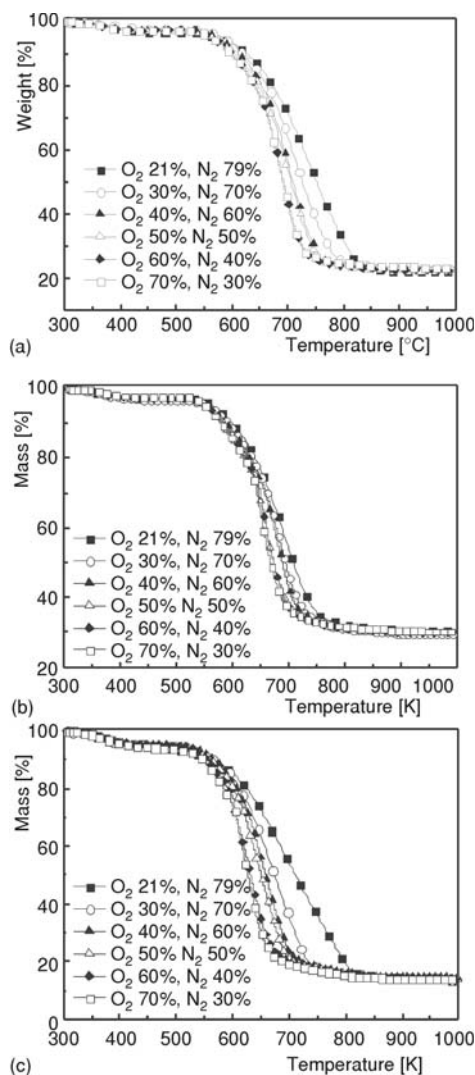


Figure 5. TG profiles of (a) PZ lignite, (b) HLH lignite, (c) ZLNE lignite

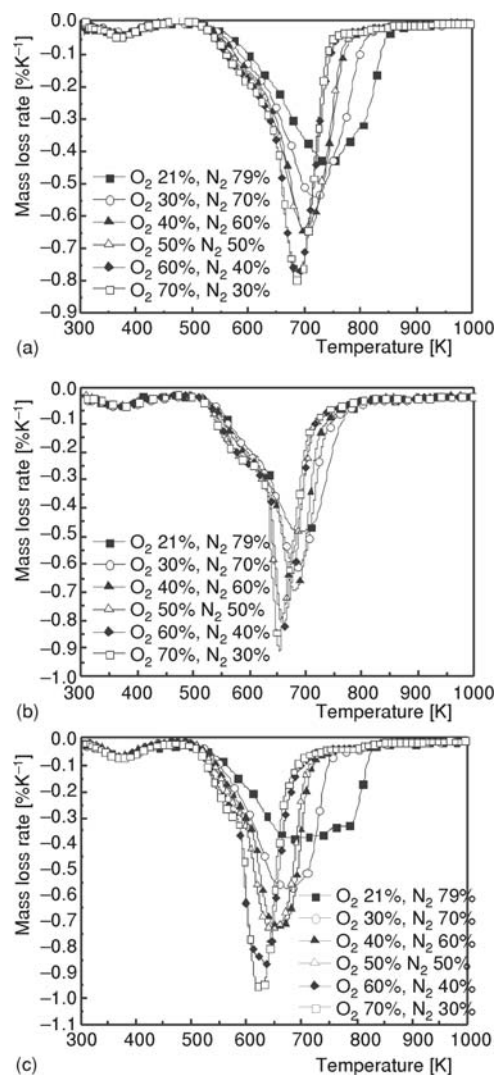


Figure 6. DTG profiles of (a) PZ lignite, (b) HLH lignite, (c) ZLNE lignite

The mass loss is not related to the oxygen concentration in the first stage, but at higher temperature zones, which is associated with large number of volatile decomposition and char burning at the second stage. The high oxygen concentration makes the TG/DTG/DSC curves shift to the low temperature zone. The reaction became intensely with the oxygen concentration increase. In the third combustion stage (the burn out temperature zone), there were no mass loss, which indicated that the third combustion stage was not affected by the oxygen concentration. The second combustion stage was discussed mainly in this paper. As seen in fig. 6, a single peak is observed in different oxygen conditions. In general, the less developed coals, such as lignite, present only one prominent peak in each DTG curve, and there was no obvious boundary between the devolatilization and carbon burning like bitumite.

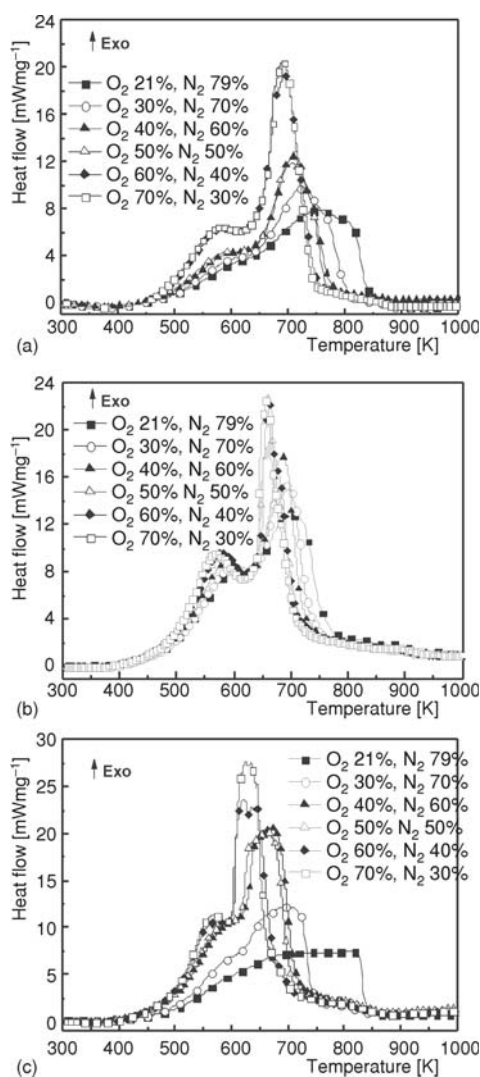


Figure 7. DSC profiles of (a) PZ lignite, (b) HLH lignite, (c) ZLNE lignite

The PZ lignite and the ZLNE lignite had the same change pattern. The characteristic temperature changed between 21-40% and 50-60% oxygen; when the oxygen concentration was above 60%, the influence of the oxygen level was reduced. The characteristic temperature of HLH lignite changed between 21-30% and 40-50% oxygen; when the oxygen concentration above 50%, the influence of the oxygen level was reduced. In general, the value of the characteristic temperature is not linear with the oxygen concentration.

From the combustion characteristic parameters listed in tab. 2, it can be seen that a higher O₂ concentration depressed all T_{ig} , T_b , T_{max} , and T_c , especially the burn out (T_b) temperature. The tendency with the influence of O₂ concentration increasing on the PZ lignite, HLH lignite, and ZLNE lignite were nearly the same. While the difference between them have been re-

Two exothermic peaks in fig.7 shifted to lower temperatures and became narrow with increased oxygen content. The demarcation point of the two peaks was more obvious in the presence with the high oxygen content than at a low oxygen level because the higher oxygen level enhanced the volatile compound burning rate, and the faster combustion reaction is not enough to ignite the residual volatile compounds and carbon at that temperature, which makes the reaction trail off for a longer time. The first exothermic peak was due to the most volatile decomposed, and the second exothermic peak was due to the combustion of the carbon and the residual volatile decompose. The second exothermic peak was sharper than the first one because the carbon combustion reaction is more intense than the volatile decompose.

Oxygen-enriched combustion characteristic parameters

Characteristic temperature

The characteristic temperatures were shown in tab. 2. From the characteristic temperatures in tab. 2, it can be observed that the value of T_i changed lightly in each atmosphere for the three types of raw lignite, which indicates that the initial volatile evolution of lignite is not or weakly affected by the oxygen concentration. The initial decomposition process only related to the temperature. Values of T_{ig} , T_c , T_{max} , and T_b decreased with the increased oxygen concentration. The effect of the oxygen concentration on the peak temperature and burn out temperature were the most remarkable. The PZ lignite and

flected. The burn out temperature decrease more quickly for PZ lignite and ZLNE lignite with higher O_2 concentration. The variation of HLH lignite was relatively small. The ignition temperature (T_{ig}) was lower of ZLNE lignite than the other lignite which was attribute to its higher volatile concentrate. The T_{max} and T_c point of PZ lignite were relatively higher than the other lignite. The above difference indicated that the higher calorific value the greater influenced by the oxygen concentration of the burning rate.

Table 2. The value of characteristic temperature

	$O_2:N_2$	T_i [K]	T_{ig} [K]	T_c [K]	T_{max} [K]	T_b [K]
PZ	21:79	430.78	602.14	635.94	742.46	835.84
	30:70	428.59	585.15	630.35	718.69	796.75
	40:60	430.39	577.72	623.83	705.58	770.28
	50:50	427.97	571.51	623.83	700.26	765.35
	60:40	426.30	563.22	620.71	687.77	746.58
	70:30	426.11	559.93	620.70	687.16	739.60
HLH	21:79	425.23	565.39	628.63	693.37	761.13
	30:70	428.97	561.59	627.33	685.38	744.87
	40:60	431.46	556.52	625.56	681.76	734.57
	50:50	427.61	549.44	618.20	655.65	719.96
	60:40	427.95	546.38	617.95	662.15	719.97
	70:30	427.98	541.58	617.02	654.26	711.05
ZLNE	21:79	433.91	566.54	636.81	705.23	817.16
	30:70	426.37	553.56	621.49	678.73	746.86
	40:60	425.49	546.34	610.71	657.75	720.45
	50:50	432.11	542.43	606.01	650.10	712.78
	60:40	428.69	532.07	594.91	628.99	698.86
	70:30	427.98	529.48	593.95	625.72	689.84

It can be confirmed that the oxygen level primarily influences the second combustion stage, from the ignition point to the burn out point. It took a shorter time in higher oxygen concentration environment, which improved the value of CVT. The results are presented in tab. 3.

Table 3. The burning time and the calorific value in unit time

Proportion		$O_2:N_2$					
		21:79	30:70	40:60	50:50	60:40	70:30
Burning time [min]	PZ	23.37	21.16	19.26	19.38	18.34	17.97
	HLH	19.57	18.33	17.81	17.05	17.36	16.95
	ZLNE	25.06	19.33	17.41	17.04	16.68	16.04
CVT [MJkg ⁻¹ min ⁻¹]	PZ	0.738	0.816	0.897	0.891	0.941	0.961
	HLH	0.640	0.684	0.704	0.735	0.722	0.739
	ZLNE	0.672	0.871	0.967	0.988	1.009	1.050

The non-linear fitting method was used to investigate the relationship between oxygen concentration and CVT. The fitted profiles are shown in fig. 8. The fitted results are shown in tab. 4. It is evident from tab. 4 that the correlation between the oxygen concentration and CVT fit the exponential function exactly.

Table 4. Fitted functions of the sample

Fitting function	$y = y_0 + A_1[1 - \exp(-x/t_1)] + A_2[1 - \exp(-x/t_2)]$						Chi ² /DoF	R ²
	y_0	A_1	A_2	t_1	t_2			
PZ	-0.1106	1391964.48817	0.91705	631365240.37552	10.16421	0.00087	0.97487	
HLH	0.37355	0.18364	0.18364	16.2398	16.2409	0.00026	0.96315	
ZLNE	-3.67055	4.55259	3.4361E13	7.32868	1.4835E16	0.00029	0.99694	

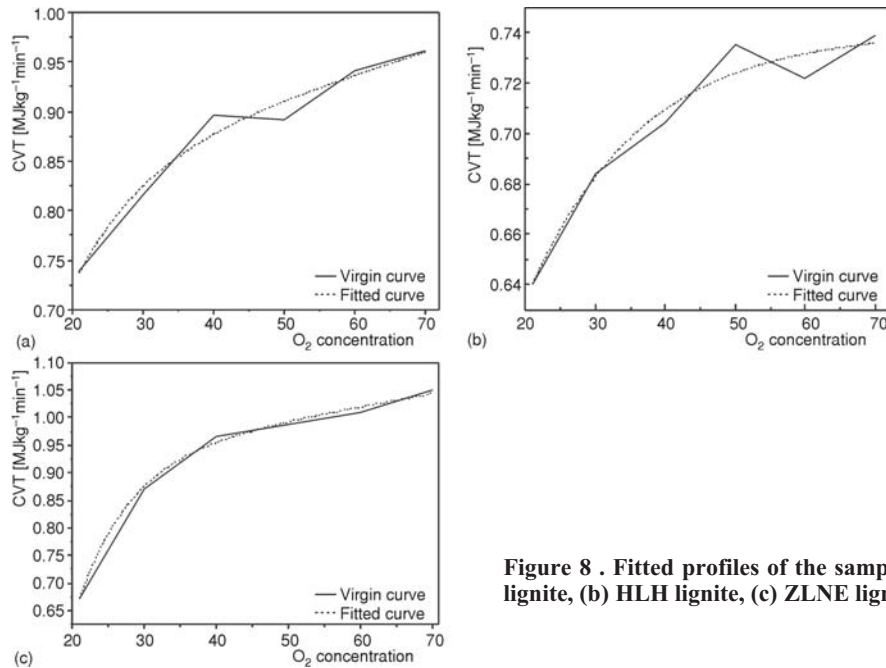


Figure 8 . Fitted profiles of the sample (a) PZ lignite, (b) HLH lignite, (c) ZLNE lignite

Combustibility index

The characteristic temperatures are combined according to the previous function to evaluate the comprehensive combustion performance. The ignition index (D'), the steady combustion index (R_w), and the combustion index (S) are calculated with the following equations.

D' is deduced from the D index, which is the ratio of $(dW/dT)_{\max}$ to T_{ig}^2 [23, 24]. It could be calculated by analyzing and comparing the flammability and the combustion reaction ability in the early stage of the combustion of the coal sample. D' is formed by adding the new characteristic value T_c to index D , and can be defined in eq. (1):

$$D' = \frac{\left(\frac{dW}{dT}\right)_{\max}}{T_c T_{\text{ig}}} \quad (1)$$

where $(dW/dT)_{\max}$ is the maximum rate of mass loss. D' represents the ignitability and burning capacity in the early stage of the lignite. From the explanation, the larger the D' value, the more volatile compounds separated from coal sample, so combustion occurs easily in the early stage.

R_w is used to indicate the combustion stability [25]. The index is defined in eq. (2):

$$R_w = \frac{560}{T_{ig}} + \frac{650}{T_{\max}} + 0.27 \left(\frac{dW}{dT} \right)_{\max} \quad (2)$$

The larger the value of R_w , the more stable of the combustion action is.

The index D' , which is a good reflection of the ignition and combustion ability in the early stage, and R_w indicates the stability during burning, but does not reveal the combustion activity in general. Thus, another index S is defined in eq. 3 [23, 25] which is presented in related literatures:

$$S = \frac{\left(\frac{dW}{dT} \right)_{\max} W_{kr}}{T_{ig}^2 T_b \tau} \quad (3)$$

where W_{kr} is the mass of the sample excluding the moisture and ash, and τ – the time of sample combustion.

This index S , which encompasses the ignition temperature, the combustion rate and the burn out temperature, is a comprehensive parameter used to compare the oxygen-enriched combustion performance of lignite. The larger the value of the index S , the higher combustion activity is. Each combustion index is presented in tab. 5.

Table 5. Combustion index in different oxygen concentrations

	O ₂ :N ₂	D'/(·10 ⁻⁵ mgK ⁻²)	R_w	S/(·10 ⁻⁹ mg ² min ⁻¹ K ⁻⁴)
PZ	21:79	1.13	2.98	4.95
	30:70	1.46	3.32	7.44
	40:60	1.83	3.67	10.45
	50:50	1.80	3.63	10.38
	60:40	2.20	4.01	13.80
	70:30	2.31	4.11	14.89
HLH	21:79	1.37	3.24	7.69
	30:70	1.74	3.68	10.58
	40:60	1.98	3.83	12.62
	50:50	2.37	4.19	16.39
	60:40	2.45	4.23	16.52
	70:30	2.71	4.48	19.13
ZLNE	21:79	1.04	2.93	5.54
	30:70	1.66	3.51	11.83
	40:60	2.15	3.96	17.54
	50:50	2.21	4.00	18.62
	60:40	2.75	4.44	24.43
	70:30	3.03	4.67	28.23

As seen in tab. 5, the value of the combustibility indexes increased with increased oxygen concentration, which indicates that enriched-oxygen conditions can improve the lignite combustion performance. The oxygen content affects the combustibility more obviously for ZLNE lignite than PZ and HLH lignite.

Conclusions

In this research, the thermal characteristics of lignite during oxygen-enriched combustion and pyrolysis were determined by TG/DTG/DSC thermal analysis methods. Our results showed the following conclusions.

- The decomposition process of the lignite is only affected by the temperature and was not related to the volatile content.
- The higher oxygen concentration can obviously improve the burning rate of the lignite, which shortened the burning time. It can be deduced in theory that the higher oxygen concentration increased the value of calorific in per unit time of the lignite. Accordance with the requirements of the heat of calcining cement and the value of calorific in per unit time, we can choose the suitable oxygen concentration in calcining cement process. The oxygen concentration and the calorific value per unit time (CVT) exactly fit the exponential function. Which can be used it to judge the trend of the lignite combustion changing in the oxygen-enriched atmosphere.
- Not all the lignite suitable to be used as fuel for calcining cement by using improved the oxygen concentration. Compared with the high calorific lignite, the low calorific value of the lignite was affected weakly by the oxygen concentration when burning. They need the higher oxygen concentration to reach the requirement in calcining clinker. Economical and operability considerations, it was not a feasible method.
- The fixed carbon initial burning temperature was defined (T_c) as the starting point of the second exothermic peak in the DSC curve. From that the combustion process can be divided into four stages. The first stage is moisture evaporation and partial volatile decomposition. The second stage is the most volatile matter decomposition, which is from ignition point (T_{ig}) to T_c point. The third stage is the char combustion and the rest of the volatile decomposition, which is from the T_c point to the burn out point (T_b). The last stage is for burn out.

Acknowledgments

This work was financially supported by National Natural Science Foundation of China (21066009), Scientific and Technological Project of Inner Mongolia (20080404 ZD01), New Century Excellent Researcher Award Program from Ministry of Education of China (NCET:2009-09-0865) and Project of Inner Mongolia education department (NJ09071).

Nomenclature

a	– mass loss, [%]
T_b	– burn out temperature, [K]
T_c	– fixed carbon initial burning temperature, [K]
T_i	– initial decomposition temperature, [K]
T_{ig}	– ignition temperature, [K]
T_{max}	– peak temperature, [K]

Acronyms

CVT	– calorific value per unit time, [MJkg ⁻¹ min ⁻¹]
DSC	– differential scanning calorimetry, [mWmg ⁻¹]
DTG	– derivative thermogravimetric analysis, [%·K]
TG	– thermogravimetry, [%]

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