CHEMICAL INTERACTION EFFECTS OF CARBON DIOXIDE GASIFICATION AND OXYGEN OXIDATION ON OXY-CHAR BURNING RATES UNDER MODERATE AND HIGH TEMPERATURE

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Abstract Oxy-char burning is an important process for oxyfuel combustion under moderate and high temperatures. The chemical crossover interaction effects of carbon dioxide gasification and oxygen oxidation are still unclear to need further investigate. Experimental investigations were implemented on oxy- and air-char transient and mean burning rates, particle temperature and char burning rate ratio between gasification and oxidation under moderate and high temperatures. The Meng bituminous coal char burning in 27%O_2/CO_2/A_r, 27%O_2/A_r, 73%CO_2/A_r, and benchmark case 21%O_2/N_2/A_r under 1273K and 1773K, were performed in a novel oxy-combustion experiment system. Results showed that the oxy-char burning rate was distinguishably affected by chemical crossover interaction between oxygen oxidation and carbon dioxide gasification. In high temperature, controlled by chemical interaction of oxidation and gasification, the maximum burning rate ratio as 34.8% from gasification was smaller than that as 47.1% in moderate temperature due to char porous structure reduction induced by high temperature ash melting. The char mean burning rate of sole carbon dioxide gasification in 1773K was 1.25 times to that in 1273K, but burning rate under 27%O_2/CO_2/A_r was 0.69 times to that in moderate temperature. Burning rate ratio of sole carbon dioxide gasification increased from 0.28 to 0.40 correspondingly in moderate and high temperatures. Char burning temperatures for serious melting combustion were generally identical and even flame distribution in char surface layer illuminated as luminous flameless combustion.

1 INTRODUCTION

Oxyfuel combustion is a novel low-carbon technology to develop clean pulverized coal power plant, i.e., PC, and also utilize in the effective coal gasification including the gasifier, the circulating fluidized bed, i.e., CFB, and the entrained-flow bed, i.e., EFB. (Wall et al., 2009; Toftegaard et al., 2010; Chen et al., 2012; Zheng et al., 2015) In oxyfuel combustion, the gasification of carbon dioxide, gas water and the oxidation of oxygen have complex competing crossover chemical interactions. Investigation of competing crossover chemical interactions of gasification from carbon dioxide, gas water and oxidation from oxygen is necessary to improve the char burning rates, high burnout and luminous oxy-flame propagation. (Toporov et al., 2008; Zhang et al., 2011; Heil et al., 2011; Liu et al., 2003; Glarborg et al., 2008; Liu et al., 2012; Guo et al., 2015, 2017; Chen et al., 2015)

In the past decades, investigations of gas and solid fuel oxy-combustion showed that the burning rate is generally reduced but steam addition increases burning rate under high temperature. Few investigations are carried out on the competing crossover chemical effects of carbon dioxide gasification and oxygen oxidation on the coal char burning rates, particles temperature, and flame propagation. (Zhou et al., 2016; Roy et al., 2016; Liu et al., 2012, 2015, 2017; Kim et al., 2014;) Especially, char burning rate ratio from endothermic gasification of carbon dioxide is still unclear and need further distinguish from the total crossover chemical reactions, i.e., the carbon dioxide endothermic gasification and oxygen exothermic oxidation, respectively. In previous literature, researchers revealed the oxy-char distinguished burning characteristics due to the different physicochemical properties, i.e., thermophysical and chemical properties of carbon dioxide and steam in dry and wet recycled flue gas oxyfuel combustion. Zhou et al. (2016) researched on the relative contributions of oxygen concentration, thermal and chemical effects on the char combustion rate at an ambient gas temperature of 1200K for bituminous coal char with its mean diameter as 91μm. Kim et al. (2014) and Hecht et al. (2011, 2012, 2013) revealed that in low oxygen concentration the carbon dioxide gasification increases the overall char consumption rate but in high oxygen concentration its gasification decreases char consumption rate. Gonzalo-Tirado et al. (2012) numerically illuminated the overall char consumption rate using a single film model from 4-21 vol. % in N_2 and CO_2 at 1573K and showed that carbon dioxide gasification consistently enhances the overall char conversion rate. Zhang and Binner et al.

(2010) found that the coal char burning temperature is lower in O_2/CO_2 atmosphere and char gasification with carbon dioxide improves the coal burning rate.

Present work mainly emphasizes on the chemical interaction effects of carbon dioxide gasification and oxygen oxidation on oxy-char burning rates under moderate and high temperatures. Purpose is to understand the carbon dioxide gasification increasing and decreasing char burning rates. Especially, the char burning ratio from carbon dioxide endothermic gasification is distinguished from exothermic oxygen oxidation under moderate and high temperatures atmospheres, i.e., 1273K and 1773K, which are suitable to the oxy-CFB and oxy-PC combustion especially with fusion occurring, respectively. This work helps improve oxyfuel combustion industrial utilization and benefit char burnout model development.

2 EXPERIMENTS

2.1 Experiment setup

The oxyfuel combustion experimental system consists of the $O_2/CO_2/N_2/A_r$ gas mixture unit, the vertical chemical reactor, the fuel sample carrier, the gas and particle temperature measurement, the digit camera for capturing flame structure, the data sampling and reserving computer, shown in Figure 1. In the inner middle of furnace, the coal char in reaction tube is heated by the electric heating equipment, which mainly is a type of high quality pure alumina corundum with high resistance temperature as 1973K.

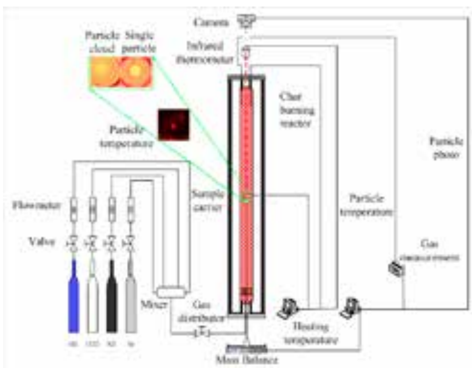


Fig. 1. A schematic diagram of oxyfuel combustion system

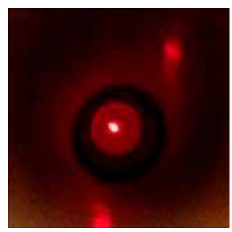


Fig. 2. The measurement position using infrared thermometer in the middle of tube

2.2 Fuel characteristics and experimental parameters

In the experiment, the Meng bituminous coal was used and its proximate and ultimate analysis showed in Table 1. For obtaining the effects of carbon dioxide gasification on the char burning rates under moderate and high temperatures, two different temperatures of 1273K and 1773K and the coal char diameter range as 90-125μm were chosen in six oxyfuel combustion cases. High temperature of 1773K was chosen to investigate coal char combustion characteristics with ash melting suitable to high temperature coal combustion in pulverized coal power plant. Moderate temperature of 1273K was benchmark experimental atmosphere and also comparatively suitable to oxyfuel circulating fluidized bed (as 1073~1273K). Six oxy-char burning experiments were 1273K 27%$O_2/CO_2/A_r$; 1773K 27%$O_2/CO_2/A_r$; 1273K 27%O_2/A_r; 1773K 27%O_2/A_r; 1273K 73%CO_2/A_r; 1773K 73%CO_2/A_r. Two benchmark experiments of 1273K 21%$O_2/N_2/A_r$ and 1773K 21%$O_2/N_2/A_r$ were also investigated to compare with oxy-char burning characteristics. Generally, the 27% volume fraction of oxygen in oxyfuel combustion brings the heat transferring flux ratio between convection and radiation heat transfer to be similar with that in air combustion for pulverized coal power plant.

Table 1.Meng Coal Proximate and Ultimate Analysis（% ar）[a]

VM	FC	M	A	C	H	N	S	O	QLCV
22.70	34.09	14.87	28.35	43.70	2.80	0.71	0.70	8.87	16.39

[a] VM = volatile matter, FC = fixed carbon, M = moisture, A = ash, LCV = low calorific value (unit: MJ/kg).

In experimental process, the coal char burning with high temperatures involved the coal ash melting, and its mineral components are shown in Table 2. In the moderate temperature of 1273K, none fusion occurred in char burning reaction. High temperature of 1773K beyond coal ash melting temperature, i.e., flowing temperature of 1703K, yielded a char burning in fusion state, which involved relatively different physical char structure, burning rate and chemical reaction.

Table. 2: Meng Coal Ash Minerals Properties （%）

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO
35.04	39.02	3.90	9.82	0.35
TiO_2	SO_3	P_2O_5	K_2O	Na_2O
1.78	5.86	0.21	0.40	0.28

Experimental cases and operation parameters for char burning under $O_2/CO_2/N_2/A_r$ atmosphere are shown in Table 4. The flow volume rate was 0.40 l/min in char burning experiments. Total experiments included six oxy-char burning experiments and two air-char burning benchmark experiments in the moderate temperature of 1273K and the high temperature of 1773K, respectively. The additional gas species argon, i.e., A_r, was used as the equilibrium carrier gas for experimental cases.

Table. 4: Experimental properties and parameters for char burning under $O_2/CO_2/N_2/A_r$ atmosphere

	Oxy-char			Air-char
Index	a	b	c	d
Experimental cases	$27\%O_2/CO_2/A_r$	$27\%O_2/A_r$	$73\%CO_2/A_r$	$21\%O_2/N_2/A_r$
Temperature /K		1273		1273
Char initial mass (m±Δm) /g	5.0-0.76	5.0-0.14	5.0-0.56	5.0-0.41
Index	e	f	g	h
Experimental cases	$27\%O_2/CO_2/A_r$	$27\%O_2/A_r$	$73\%CO_2/A_r$	$21\%O_2/N_2/A_r$
Temperature /K		1773		1773
Char initial mass (m±Δm) /g	5.00+0.00	5.00+0.10	5.00+0.07	5.00+0.01
O_2 volume rate / l/min	0.108	0.108	-	0.084
CO_2 volume rate / l/min	0.292	-	0.292	0.316
A_r volume rate	-	0.292	0.108	-
Total Volume / l/min		0.40		

3 RESULTS AND DISCUSSION

3.1 Oxy- and air-char transient burning rates

The Meng coal oxy-char and air-char transient burning rates with time changes for 1273K and 1773K are shown in Figure 3 and Figure 4. The oxy-char burning rates in high temperature of 1773K were respectively higher than the corresponding char burning rates in moderate temperature of 1273K. Result shows that with the temperature increasing, the carbon dioxide gasification reaction increases and the oxy-char burning rate also improves as well as oxygen oxidation. In the moderate temperature of 1273K, the oxy-char burning rate of $27\%O_2/CO_2/A_r$ was closely similar with the air-char burning rate of $21\%O_2/N_2/A_r$, corresponding to char maximum burning rate as about 0.26 g/min. The burning times for both cases were almost the same under moderate temperature combustion. But in high temperature, the burning times were different due to char melting.

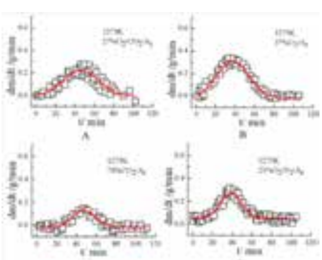


Fig. 3. Meng coal char transient burning rates in

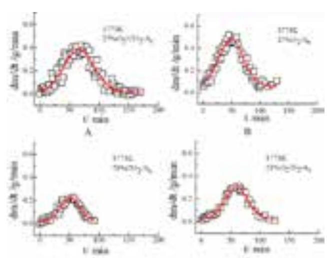


Fig. 4. Meng coal char transient burning rate in

1273K (char particles diameter: 90-125μm, coal char: 1073K, 1h; A. 27%O_2/73%CO_2/A_r; B. 27%O_2/73%A_r; C. 73%CO_2/27%A_r; D. 21%O_2/79%N_2/A_r)

1773K (char particles diameter: 90-125μm, coal char: 1073K, 1h; A. 27%O_2/73%CO_2/A_r; B. 27%O_2/73%A_r; C. 73%CO_2/27%A_r; D. 21%O_2/79%N_2/A_r)

3.2 Maximum burning rates, the burning rate ratio between oxidation and gasification

The maximum burning rates illuminated the effects of temperature changes on coal char burning physicochemistry, shown in Figure 5 and Figure 6. Combustion temperature is important for controlling the pulverized coal burning rates especially in high Reynolds number turbulent combustion boiler. Figure 5 and Figure 6 show that the maximum burning rates under high temperature are normally higher than that under moderate temperature, respectively. Compared with the maximum burning rate of the 27%O_2/CO_2/A_r experiment in moderate temperature, the maximum burning rate increased 1.74 times in high temperature and the effect of gasification reaction on char burning was also improved. The maximum burning rate of gasification reaction in 1773K, i.e., 73%CO_2/A_r, was 1.72 times to that in 1273K. The maximum burning rate for 21%O_2/N_2/A_r in moderate temperature was about 1.10 times than that in high temperature due to high temperature char burning, i.e., 1773K, with serious ash melting and reduction of pore structure and specific surface area. Compared with the maximum burning rate increment of 21%O_2/N_2/A_r, the maximum burning rate increment of 27%O_2/CO_2/A_r was larger because in high temperature the gasification effect of carbon dioxide increased the char burning reaction rate and improved the char particle pore structure and specific surface area for benefiting oxygen permeation within char particle and quick chemical reaction.

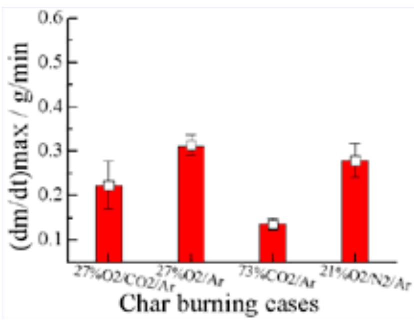


Fig. 5. Meng coal char maximum burning rates in 1273K

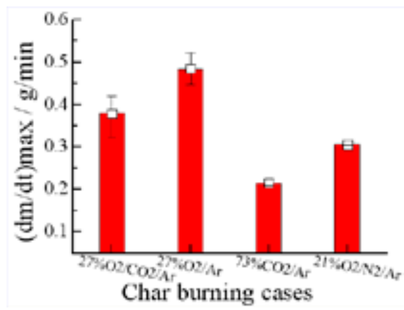


Fig. 6. Meng coal char maximum burning rates in 1773K

Nevertheless, in high temperature, the maximum burning rate ratio as 34.8% from gasification was smaller than that as 47.1% in moderate temperature, shown in Figure 7 and Figure 8. Due to distinguishing the relative contribution to the char burning rate, the char burning reaction consumption rates from oxygen oxidation, i.e., 27%O_2/A_r and carbon dioxide gasification, i.e., 73%CO_2/A_r, needed to evaluate using the coal char maximum burning ratio, respectively. The coal char maximum burning rate ratio for gasification reaction refers to the ratio of the maximum burning rate of gasification, i.e., 73%CO_2/A_r, to the maximum burning rate of oxyfuel crossover reaction, i.e., 27%O_2/CO_2/A_r. The char maximum burning rate ratio for oxidation reaction refers to the ratio of the maximum burning rate of oxidation, i.e., 27%O_2/A_r, to the maximum burning rate of crossover reaction, i.e., 27%O_2/CO_2/A_r as follows

$$RR_{CO2,max} = (R_{O2,max}-R_{O2/CO2,max})/(R_{O2,max}-R_{CO2,max}) \quad (1)$$

$$RR_{O2,max} = (R_{O2/CO2,max}-R_{CO2,max})/(R_{O2,max}-R_{CO2,max}) \quad (2)$$

in which, the gasification maximum burning rate ratio $RR_{CO2,max}$, the gasification maximum burning rate $R_{CO2,max}$, the oxidation maximum burning rate ratio $RR_{O2,max}$, the oxidation maximum burning rate $R_{O2,max}$.

Results illuminated that the gasification rate ratio of carbon dioxide decreased about 12.3% with the temperature increasing to 1.39 times due to ash melting in high temperature. With temperature increasing, the oxy-char burning reaction rates, i.e., 27%O_2/CO_2/A_r, 27%O_2/A_r, 73%CO_2/A_r, were improved, respectively. The oxy-char burning rate from chemical crossover reaction, i.e., 27%O_2/CO_2/A_r, increased about 1.74 in high temperature, shown in Figure 6. Compared with the oxy-char burning rate from carbon dioxide gasification in moderate temperature, this burning rate from gasification increased even though its burning rate ratio decreased in high temperature.

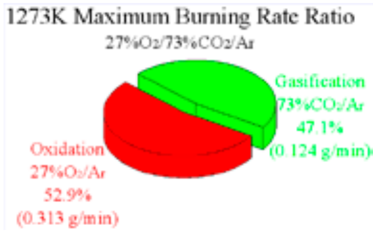


Fig. 7. Meng coal char maximum burning rate ratio for oxidation and gasification in 1273K

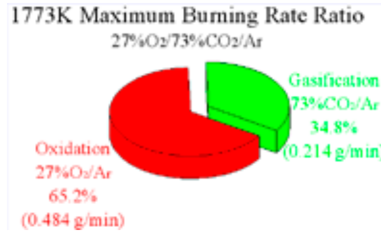


Fig. 8. Meng coal char maximum burning rate ratio for oxidation and gasification in 1773K

3.3 Temperature for maximum burning rate

Figure 9 and Figure 10 show that the coal char burning temperature changes with the different gas mixture under moderate and high temperatures. Compared to the moderate temperature, the char burning temperature was obviously higher in high temperature. Compared with the surface burning temperature in 27%O_2/CO_2/A_r, the temperature reduced about 273K in 73%CO_2/A_r due to the carbon dioxide gasification effects. However, with temperature increasing, the carbon dioxide gasification effects were larger for oxyfuel combustion. Under moderate temperature, the coal char burning temperature from gasification in 73%CO_2/A_r was nearly similar with other atmospheres. The main reason is that the physical effect of carbon dioxide, i.e., high specific heat capacity, relatively enhances in moderate temperature combustion.

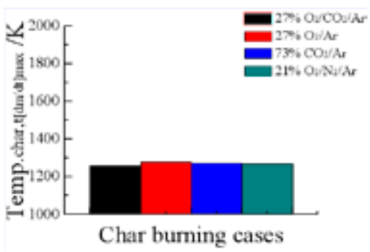


Fig. 9. Meng coal char temperature to maximum burning rate in 1273K

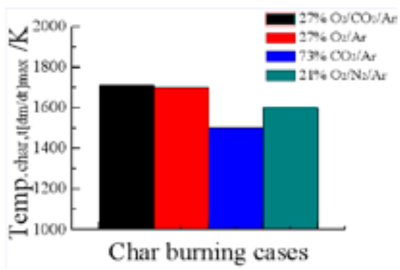
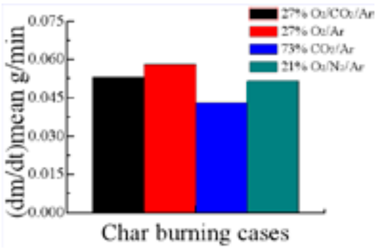


Fig. 10. Meng coal char temperature to maximum burning rate in 1773K

3.4 Mean burning rate, burnout time, ash fusion

Figure 11 and Figure 12 show the char mean burning rate changes with char burning atmospheres under different temperatures in reaction tube. Combustion temperature turning from moderate to high temperature brought drastically changes in char burning rate mainly due to the crystal phase transporting from solid phase without fusion to fluid phase with fusion in char burning. The results showed that in high temperature, the combustion reaction with fusion brought the lower char burning rate except for the gasification reaction, i.e., 73%CO_2/A_r. The main reason is that the porous char reaction temperature was lower for endothermal gasification reaction and slight fusion. All these caused char surface kinetics reaction rate contrarily increased under certain gas diffusion. In high temperature, the char mean burning rate of gasification reaction, i.e., 73%CO_2/A_r, was 1.25 times to that in moderate temperature. Contrarily, in the high temperature, this burning rate from 27%O_2/CO_2/A_r was 0.69 times to that in moderate temperature. It illuminates that carbon dioxide gasification for pulverized coal char is effective to improve char burning rate in high temperature.



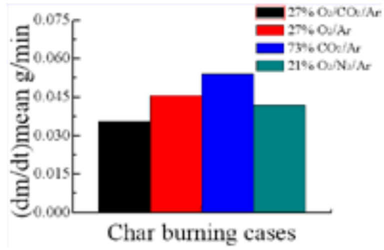


Fig. 11. Meng coal char mean burning rates in 1273K

Fig. 12. Meng coal char mean burning rates in 1773K

In char burning, ash fusion was important to determine the char burning rate, carbon dioxide gasification effect and chemiluminesence laminar flame propagation, including char surface temperature, burnout time, physical structure and gaseous emissions, etc. Figure 13 shows that oxy- and air-char burning rates are distinguished by char ash fusion and laminar luminous burning flame propagation. In moderate temperature, the burned char fragments contributed to the char burning reaction without slight or serious fusion. The fragments of char ash had infinitely small pores. These pores were less than 0.1mm and mostly produced by carbon dioxide gasification reaction. Especially, in high temperature, the fragments had absolutely diminished and transformed into a absolute char fusion structure which yielded flowing thermal bubbles illuminated as serious fusion, i.e., 1773K 27%O_2/CO_2/A_r, 1773K 21%O_2/N_2/A_r, and even infinitely spherical small particles or balls in slight fusion, i.e., 1773K 27%O_2/A_r, 1773K 73%CO_2/A_r.

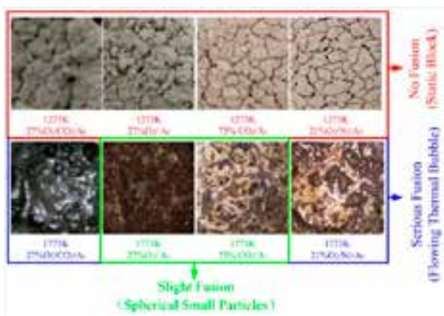


Fig. 13. Meng coal char burning photos in 1273K and 1773K (meltingindex: DT 1663K, ST1683K, HT 1693K, FT1703K)

4 CONCLUSIONS

Under moderate and high temperatures, compared with benchmark air-char burning, i.e., 21%O_2/79%N_2/A_r, the chemical interaction effects of carbon dioxide gasification and oxygen oxidation on Meng coal char burning rates were experimentally investigated through oxidation reaction, i.e., 27%O_2/A_r, gasification reaction, i.e., 73%CO_2/A_r, and crossover overlapping competing reaction between oxidation and gasification, i.e., 27%O_2/73%CO_2/A_r, in the present work. The conclusions are as follows:

1) In moderate and high temperatures, the chemical crossover interactions of carbon dioxide gasification and oxygen oxidation had relatively distinguishably effects on coal char maximum and mean burning rates, char burning time, char burning temperature and char physical structure. In high temperature, the maximum burning rate ratio of carbon dioxide gasification as 34.8% for oxy-char burning contrarily more reduced than that in moderate temperature, i.e., 47.1%, due to ash melting combustion. Burning rate ratio of sole carbon dioxide gasification increased from 0.28 to 0.40 in moderate and high temperatures.

2) Char mean burning rates of sole oxidation, i.e., 27%O_2/A_r, sole gasification, i.e., 73%CO_2/A_r, and crossover reaction, i.e., 27%O_2/CO_2/A_r, varied with char burning atmosphere temperature. In high temperature, Ash melting and covering pores structure occurred in oxy-char burning which reduced the coal char mean burning rates. This mean burning rate of sole carbon dioxide gasification in 1773K was 1.25 times

to that in 1273K, but this mean burning rate for crossover reaction was 0.69 times to that in moderate temperature due to high temperature ash melting.

3) Char burning temperature was almost the same with the atmosphere temperature in moderate temperature combustion, but in high temperature the endothermic gasification absorbing char surface chemical heat acquired lower burning surface layer temperature as decreasing about 200K. Oxy-char mean burning rate was nearly the same but the maximum rate was lower as about 20% decreasing. In the high temperature of 1773K, this mean rate was lower just about 15% decreasing but the maximum rate was higher just as about 25% increasing.

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