INVESTIGATION OF THE POLLUTANT EMISSION CHARACTERISTICS OF BLENDS OF BIOMASS AND COAL GANGUE IN A FLUIDIZED BED

by

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The composite fuel of coal gangue and biomass is expected to increase the utilization rate of solid waste and compensate for the disadvantages of separate fuels. However, the NO and SO_2 emissions from this composite fuel during the combustion process are a concern, but there are few studies on it. In this paper, corn cobs and wheat straws, typical agricultural biomass discarded in North China, and coal gangue from the mine in Xilingol, China, were selected for co-combustion in a fluidized bed. The emission characteristics of NO and SO_2 were studied by changing the ratio of biomass to coal gangue and the combustion temperature. Studies had shown that: to a certain extent, mixing biomass, and coal gangue can reduce the NO and SO_2 emissions, and mixing 20% of biomasses had the best effect on NO and SO_2 emissions reduction. The SO_2 emission reduction effect of wheat straws was better than that of corn cobs, and the NO emission reduction effect of corn cobs was better than that of wheat straws. The NO emissions of wheat straws and corn cobs added to the coal gangue were 6.45% and 7.93% less than those of coal gangue alone. The SO_2 emissions of wheat straws and corn cobs added to the coal gangue were 27.45% and 25.94% less than those of coal gangue alone. Both NO and SO_2 emissions decreased with the growth of the biomass rate; NO and SO_2 emissions increased with the growth of the combustion temperature.

Keywords*: biomass, coal gangue, co-combustion, NO, SO_2*

Introduction

Energy is a necessity for human survival and development. With the development of society, human demand for energy is increasing. Therefore, the development and utilization of renewable energy is particularly important [1]. Biomass, as a clean energy source, has received widespread attention. It has the advantages of large total amount, wide sources and low pollution, but the annual utilization rate is less than 1% of production [2]. Relevant data showed that China's raw coal output in 2019 was 3.97 billion tons, and the coal gangue (CG) produced during raw coal mining and coal washing would be more than 300 million tons [3].

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But CG is not well utilized. The unreasonable use of CG has caused great harm to human society: the harmful gases produced by its combustion were emitted into the atmosphere, polluting the environment, and the unreasonable stacking has caused many collapses [4-7]. At present, CG is mainly used in materials, construction, agriculture and energy [8]. Utilizing the complementary characteristics of CG and biomass and strengthening the comprehensive utilization of both, can not only alleviate the problem of energy shortage, but also can reduce the emission of harmful gases and pollution to the environment, reduce GHG emissions, improve the combustion efficiency, and help to build a resource-saving and environment-friendly society [9-12].

Yelvertona *et al.* [13] used various woody biomasses as substitutes for coal and studied the gas and particulate emissions of individual samples, indicating that it was reasonable to use woody biomass and coal as the alternative energy source. Sher *et al.* [14] confirmed this conclusion through experiments. Bi *et al*. [15, 16] co-fired biomass and CG in the different mass ratio, through the controlled experiment, showed that the co-combustion of the two was conducive to reducing GHG emissions, and selected the optimal fraction. The TG-DTG method and the Friedman method were used to explore the mixed combustion characteristics of biomass and CG by Zhao *et al.* [17], indicating that adding a small amount of biomass to CG can reduce activation energy and improve combustion. Gong *et al.* [18, 19] co-fired biomass and CG in the circulating fluidized bed under the oxygen-enriched atmosphere, showed that the gas emissions produced by the co-combustion of CG and biomass were related to the combustion atmosphere. Shi *et al.* [20] co-fired agricultural biomass and CG in a bubbling fluidized bed, and through the control variable method, showed that the amount of gas emissions was related to the primary air rate and the feed air rate.

Yang *et al.* [21] combined simulation and experiment, showed that the NO emissions decreased with the increase of the mixing ratio of biomass, and the addition of biomass to the coal can suppress the NO release. However, as the temperature increased, NO emissions also increased. Rokni *et al.* [22] and Yanik *et al.* [23] co-fired biomass and torrefied biomass with coal, and found that after co-firing NO emission of the raw biomass was lower than torrefied biomass. The SO_2 emission of the torrefied biomass was less than the raw biomass. This was due to the low S content of the torrefied biomass, so SO_2 emission was small. Thanet and Suneerat [24] co-fired coal and biomass in a circulating fluidized bed to study the characteristics of pollutant emissions. The results showed that when the excess air ratio was too low the NO_x emission was lower, this was because the C content was lower; while SO_2 emission was related to temperature, and SO_2 emission decreased with the increase of the excess air ratio.

In this paper, biomass and CG were co-fired in a fluidized bed, and the mass ratio of the two and the combustion temperature were controlled to investigate the pollutant emission characteristics, and then provide some references for further research.

Material and methods

Materials

In this work, corn cobs (CC) and wheat straws (WS) samples were collected from North China. The CG samples were obtained from the mine in Xilingol, Inner Mongolia, China. Referring to the relevant standard methods of the American Society for Testing Materials (ASTM D197-87, 2012) [25], the test materials were dried in the oven (202-00T LI-CHEN, China) at 105 °C for 12 hours. After preliminary drying, the test materials were ground. Biomasses were sieved to less than 100 mesh (<0.15 mm), and CG were sieved to less than 120 mesh (<0.125 mm). In order to ensure that biomass and CG had the same composition under different particle sizes, all materials on the sieve were broken several times and all were sieved. The proximate analyser (SDLA718 SUNDY, China) and the ultimate analyser (SDCHN SUNDY, China) were both used to identify and analyse the dried CG, WS, and CC. The proximate analysis and ultimate analysis were shown in tab. 1.

Table 1. Proximate and ultimate analysis of samples

Samples	Proximate Analysis [wt.%]					Ultimate Analysis [wt.%]				
	M_{ad} [%]	A_{ad} [%]	V_{ad} [%]	FC_{ad} [%]	Qb_{ad} [kJkg^{-1}]	C_{ad} [%]	H_{ad} [%]	O_{ad} [%]	N_{ad} [%]	S_{ad} [%]
CG	0.81	53.61	12.89	32.69	11386	34.07	1.37	8.64	0.41	1.09
WS	4.21	6.43	69.47	19.89	14673	44.21	4.63	39.63	0.76	0.13
CC	9.86	3.82	72.59	13.73	16216	43.05	5.18	37.42	0.59	0.08

Methods

In this experiment, the separate combustion of CG and the co-combustion of biomass and CG were carried out in a self-made 36 kW per hours small fluidized bed. The furnace chamber was cleaned with primary air before the test started. In the test, the combustion atmosphere was air. The biomass mixing ratio and the combustion temperature were varied to study NO emissions and SO_2 emissions during the combustion of CG alone and the co-combustion of CG and biomass with the same primary air volume, secondary air volume and feed air volume. The test method was shown in tab. 2.

Table 2. The scheme of fluidized bed combustion test under different ratio

Samples	Primary air volume [mgm^{-3}]	Secondary air volume [mgm^{-3}]	Feed air volume [mgm^{-3}]	Burning temperature [°C]
CG	1.8	1.5	1.0	850
CG: WS/CC = 6:4, 4:6, 2:8,8:2	1.8	1.5	1.0	850

As shown in tab. 2, when studying the effect of the mixing ratio on the pollutant emissions, the test conditions were: the primary air-flow of 1.8 mg/m^3, the secondary air-flow of 1.5 mg/m^3, the feed air-flow of 1.0 mg/m^3, and the combustion temperature of 850 °C. A total of nine groups of samples were available: CG, CG and 20% of WS, CG and 40% of WS, CG and 60% of WS, CG and 80% of WS, CG and 20% of CC, CG and 40% of CC, CG and 60% of CC, CG and 80% of CC.

Table 3. The scheme of fluidized bed combustion test at different temperatures

Samples	Primary air volume [mgm^{-3}]	Secondary air volume [mgm^{-3}]	Feed air volume [mgm^{-3}]	Burning temperature [°C]
CG	1.8	1.5	1.0	800,850,900,950
CG: WS/CC = 8:2	1.8	1.5	1.0	800,850,900,950

As shown in tab. 3, when studying the influence of combustion temperature on pollutant emissions, the working conditions were: primary air-flow of 1.8 mg/m^3, secondary air-flow of 1.5 mg/m^3, feed air-flow of 1.0 mg/m^3, combustion temperature of 800 °C, 850 °C, 900 °C, 950 °C. Three sets of samples were available: CG, CG and 20% of WS, CG and 20%

of CC. The study by Yang *et al.* [26] had showed that the best synergistic effect was obtained when the ratio of biomass to CG was 2:8 in the mixed combustion test.

The schematic diagram of the specific set-up of the fluidized bed test bench is shown in fig. 1.

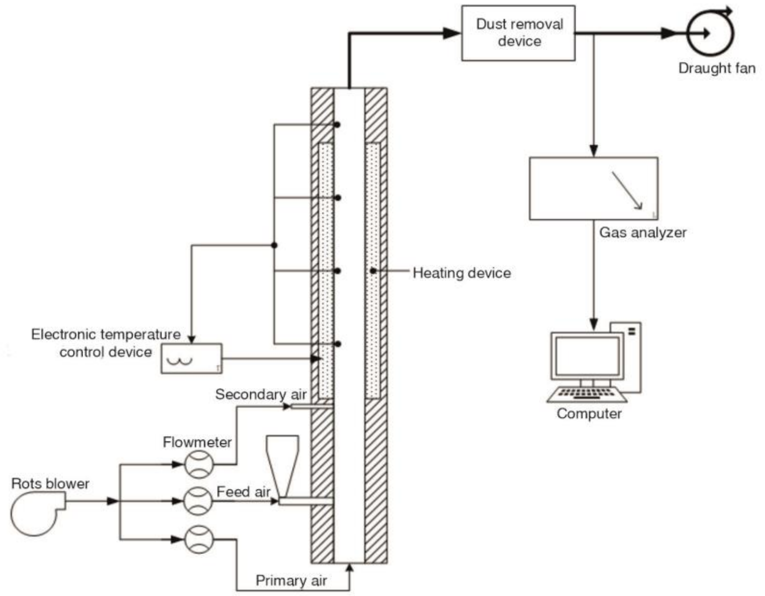

Figure 1. The schematic diagram of the fluidized bed

Results and discussion

The influence of different blending ratios on the emission characteristics of NO and SO_2

Figures 2(a) and 2(b) showed curves of NO emissions when mixing WS, CC, and CG in different proportions, respectively. It can be seen from the figures that with the increase of the mixing ratio of WS and CC, NO emissions tended to decrease. When biomasses were burnt in the upper layer of the dense phase zone and the dilute phase zone of the fluidized bed, a large amount of oxygen was consumed, forming the fuel-rich atmosphere and reducing part of NO, leading to the reduction in NO production.

For the convenience of analysis, it was assumed that the theoretical emission value for the composite fuel were the linear superposition of the CG and WS/CC emission values, which were calculated:

$$[\mathrm{NO}]_{\mathrm{theoretical}} = x[\mathrm{NO}]_{CG} + y[\mathrm{NO}]_{\mathrm{biomass}} \tag{1}$$

where x and y were the mass fractions of biomass and CG in the mixed sample, $[\mathrm{NO}]_{\mathrm{biomass}}$ – the experimental NO emission value of biomass, and $[\mathrm{NO}]_{CG}$ – the experimental emission value of CG, respectively.

The experimental emission concentration of NO and SO_2 measured in experiments were shown in fig. 3.

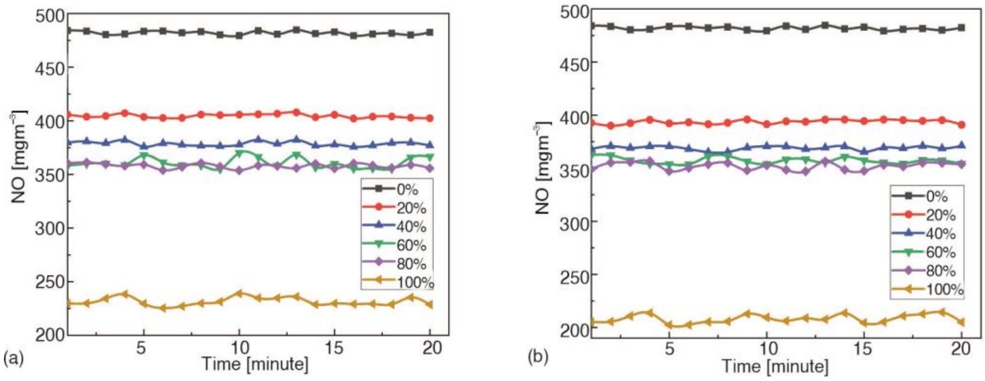

Figure 2. The NO emission curves at different mass ratios

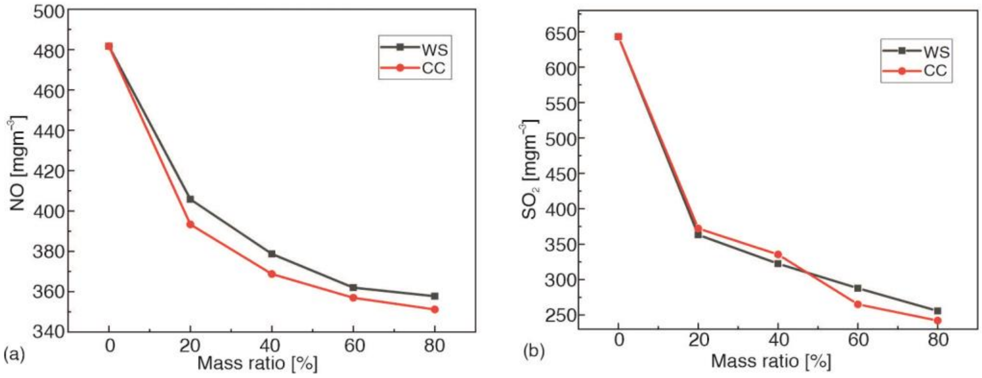

Figure 3. Effects of biomass content on NO and SO_2 emissions

Figure 3(a) presented the actual NO emission concentration. The figure illustrated that the co-combustion of biomass and CG can reduce NO emissions to a certain extent. When 20% biomass was added, NO emissions decreased rapidly, and then began to decrease slowly as the proportion of biomass blended increased. When CG was burned alone, NO emission concentration was 482 mg/m^3. After adding 20% of WS, NO emission was reduced to 404.5 mg/m^3, after adding 20% of CC, NO emission was reduced to 393.3 mg/m^3. Mixing 60% of WS and 80% of WS, NO emission concentration was basically the same, as did 60% of CC and 80% of CC. Figure 3(a) also showed that NO emissions had decreased as the proportion of WS/CC additions had increased. The difference in NO emissions from compound fuels was most pronounced with the addition of 20% WS/CC. This addition ratio contributed to a reduction in NO emissions of 6.45%/7.93% for CG, respectively. However, for the 40%/60%/80% WS/CC addition rates, the CG NO emissions also decreased slightly by 1.07%/0.98%, –0.87%/–12.43% and –23.98%/–33.61%, respectively. Proximate analyses of CG and biomass revealed that the biomass had the higher nitrogen content, but as the biomass increased, NO emission characteristic curves tended to decrease. The reason for this was that biomass consumed a large amount of oxygen during combustion, which was then distributed in the upper layer of the dense phase zone and in the dilute phase zone of the furnace. The reducing atmosphere was formed in the dilute phase zone, reducing the NO generated by the CG and biomass and forming the fuel graded combustion effect. As the mixing ratios of biomass increased NO emissions tended to be consistent. This was because the theoretical combustion air per unit mass of biomass was greater than that of CG. Under the same air-flow conditions, the local area of the furnace biomass combustion was not complete, and NO generations were reduced, resulting in the proportion of biomass increased but NO emissions did not increase. Sung *et al.* [27] also proved that NO_x would be reduced to a certain extent when biomass and coal were co-fired, and the carbon burnout rate would be increased.

Figure 3(b) showed the actual SO_2 emission concentration. It observed that after 20% of biomasses were mixed with CG, SO_2 emission concentration was greatly reduced, and SO_2 emission reduction rate decreased with the increase of mixing ratio of biomass. Moron and Rybak [28] also demonstrated that the addition of biomass can significantly reduce SO_2 emissions by co-firing biomass and coal in an oxygen-rich atmosphere. As the biomass mixing ratio increased uniformly, SO_2 emissions also decreased uniformly. During combustion, 80% of the sulphur-containing compounds in the fuel would decompose, oxidize and burn at high temperatures to produce SO_2. The proximate analysis of the test materials in tab. 1 revealed that the sulphur contents of CG were much higher than that of biomass. Therefore, adding the appropriate amount of biomass to CG can reduce the overall sulphur contents of the fuel and reduce the production of SO_2 from the source of production.

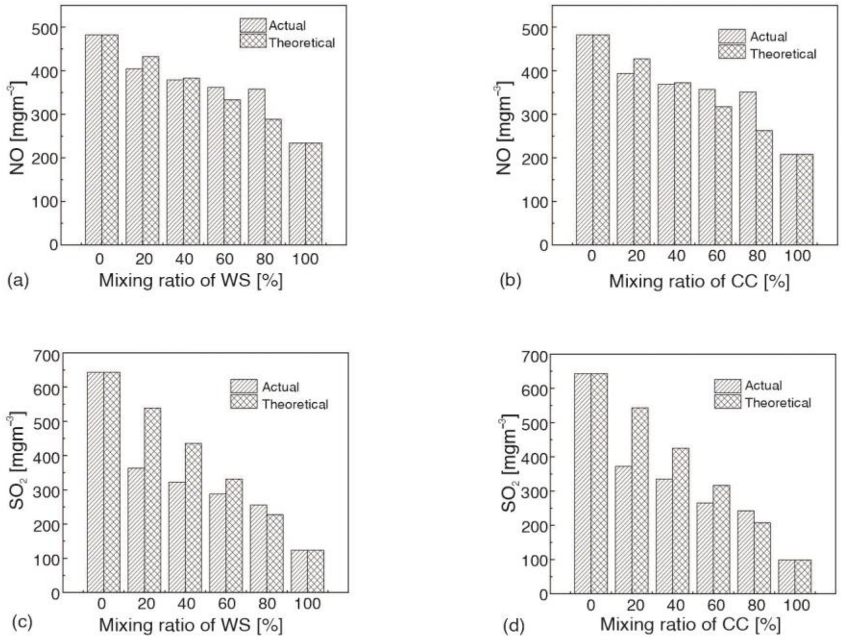

Figure 4. Comparison of theoretical and actual emissions of NO and SO_2 under different mass ratios

Figure 4 showed the comparisons of theoretical NO and SO_2 emissions and actual NO and SO_2 emissions in different mass ratios of biomass and CG. Figures 4(a) and 4(b) were comparison diagrams of theoretical and actual emissions of NO, and figs. 4(c) and 4(d) were comparison diagrams of theoretical and actual emissions of SO_2. These figures illustrated that with the increased of WS/CC, the theoretical and actual emissions had shown the downward trend. When 20% of WS/CC was blended, the difference between theoretical and actual emissions was the largest, that is, the emission reduction ratio was the largest. When 80% of

WS/CC was blended, the difference between the theoretical and actual emissions was negative, and the emission reduction effect was weakened. It can be seen from tabs. 4 and 5 that adding different proportions of biomass per unit mass of CG has different effects on NO and SO_2 emissions reduction. As the proportion of biomass blending increased, the proportion of NO and SO_2 emissions reduction gradually decreased. Both NO and SO_2 emissions reduction effects were the most obvious when 20% of biomass was added. The SO_2 emission reduction effect of WS was better than that of CC, and the NO emission reduction effect of CC was better than that of WS.

Table 4. The NO emission reduction ratio when adding biomass

Blending ratio / Biomass	0	20%	40%	60%	80%
WS	0%	6.45%	1.07%	–0.87%	–23.98%
CC	0%	7.93%	0.98%	–12.43%	–33.61%

Table 5. The SO_2 emission reduction ratio when adding biomass

Blending ratio / Biomass	0	20%	40%	60%	80%
WS	0%	32.6%	25.93%	13.15%	–12.14%
CC	0%	30.34%	21.13%	16.22%	–16.64%

The theoretical SO_2 emission concentration of burning 80% of CG alone was 514.3 mg/m^3, and the actual emission after adding biomass was 322.4 mg/m^3 and 335.4 mg/m^3. It can be seen that although a small amount of biomass was added to the fuel, and the S contents was added, but the SO_2 emission concentration at the flue gas side was reduced. The theoretical SO_2 emission concentration of burning 20% of CG alone was 128.5 mg/m^3, and the actual emission concentration after adding biomass was 255.7 mg/m^3 and 242.1 mg/m^3. At this time, the biomass mixing ratio had reached 80%, and the SO_2 generated by the oxidation of sulphur in organisms cannot be ignored.

Comprehensive analyses showed that as the mixing ratio of biomass increased, both NO and SO_2 emissions decreased to varying degrees. The decrease in NO emissions was due to the incomplete combustion of biomass in local areas in the furnace and the reduction in NO production; the decrease in SO_2 emissions was due to the reduction in the overall sulphur content of the fuel.

Influence of combustion temperature on pollutant emission characteristics

Figures 5(a) and 5(b) were the NO emission curves when 20% of WS, CC and 80% of CG were co-fired at different combustion temperatures. Figures 6(a) and 6(b) were the SO_2 emission curves when 20% of WS, CC and 80% of CG were co-fired at different combustion temperatures. From the figures, it can be seen that both NO and SO_2 emissions tended to increase with the increase of temperature. At 800 °C, the SO_2 emission of WS was greater than that of CC, which was because the S content of WS was greater than that of CC, at this time, the SO_2 conversion rate of WS was greater than that of CC. With the increase of temperature, the precipitation of inorganic sulphur and organic sulphur in CG increased, and the SO_2 emissions increased, at this time, SO_2 produced by CG accounted for the main part. At 950 °C, the difference in SO_2 produced by the combustion of the two fuels was not too great.

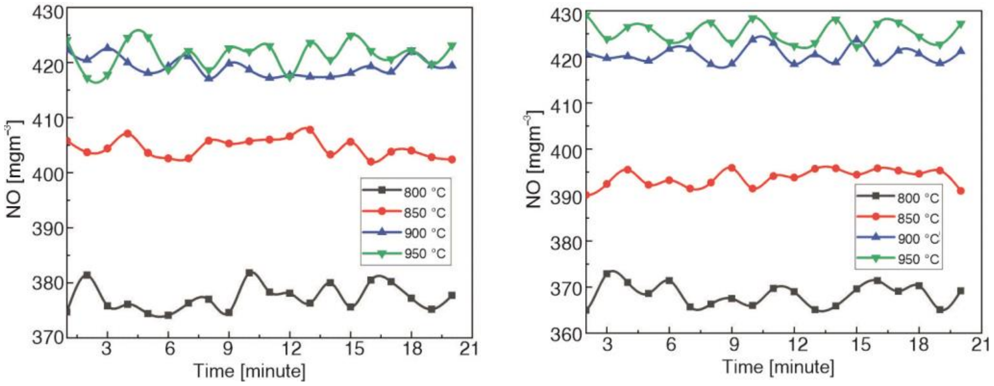

Figure 5. The NO emission curve at different combustion temperature

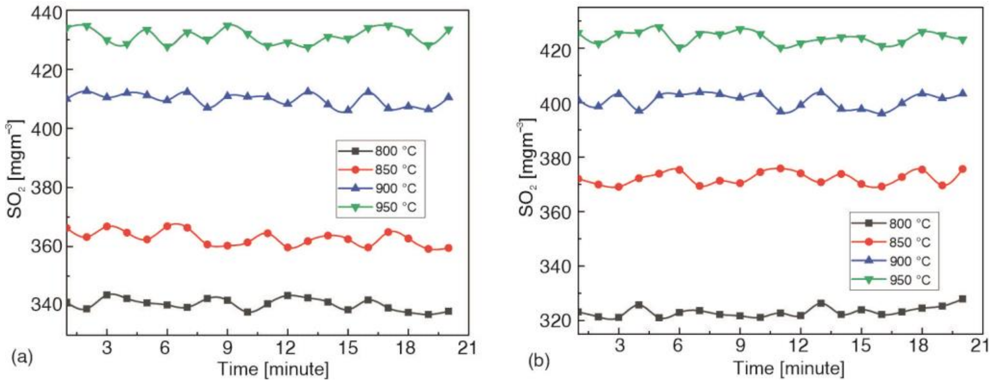

Figure 6. The SO_2 emission curve at different combustion temperature

The experimental emission concentration of NO and SO_2 measured in the experiment were shown in fig. 7. At this time, the theoretical emission value of composite fuel was also a linear superposition of CG and WS/CC emission values.

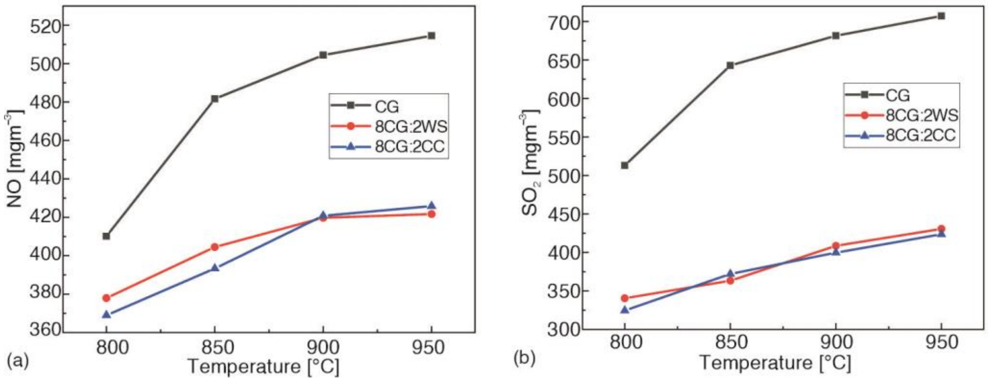

Figure 7. Effects of combustion temperatures on NO and SO_2 emissions

Figure 7(a) was the actual NO emission concentration. As can be seen from the graph, when CG was burned alone, NO emissions increased gradually with the increase of combustion temperature. The combustion temperature rose from 800 ℃ to 850 ℃, NO emissions increased from 410.1 mg/m^3 to 481.7 mg/m^3, the combustion temperature rose from 850 °C to 950 °C, NO emissions increased from 481.7 mg/m^3 to 514.5 mg/m^3. On the one hand, the increase of the combustion temperature made the precipitation of CG difficult and the precipitation of volatile matter accelerated, and the N in the volatile matter reacted to form NO. On the other hand, the nitrogen-containing compounds in coke became active with the increase of the combustion temperature. The coke violently collided and mixed in the dense phase area, and the reaction between N-containing coke and O_2 was enhanced, then NO production increased.

As indicated in fig. 7(a), when biomass was mixed with CG, NO emissions gradually increased with the increase of the combustion temperature and the increasing trend gradually slowed down. This was consistent with Wang's *et al.* conclusion [29]. This was because the volatile contents of biomass were relatively high, and the volatile nitrogen in the volatile matter was mainly released in the form of NH_3 and HCN during the combustion processed. As the combustion temperature increased, NO that produced by HCN and O_2 increased, which directly leaded to the increase of NO emissions. At the same time, coke N reacted with oxygen, and the reaction principle was the same as when CG was burned alone. Based on the above reasons, NO emissions increased with the increase of the temperature during the co-combustion of biomass and CG.

It was shown in fig. 7(b) that when the CG was burned alone, SO_2 emissions increased gradually with the increase of the combustion temperature. The combustion temperature increased from 800 °C to 850 °C and increased rapidly, and SO_2 emissions increased from 512.9 mg/m^3 to 642.9 mg/m^3, the combustion temperature increased from 850 °C to 950 °C, and SO_2 emissions increased from 642.9 mg/m^3 to 707.1 mg/m^3. As the combustion temperature increased, the precipitation of sulphur in pyrite and mercaptans and thiophenols in organic sulphur in CG accelerated, and the amount of SO_2 generated increased. The temperature continued to increase, and most of the inorganic and organic sulphur in the CG particles had been precipitated, resulting in the continued increase of the combustion temperature and the slowdown of the growth of SO_2 release, leading to continue to increase the combustion temperature, and the growth of SO_2 release slowed down. When Lupianez *et al.* [30] conducted the combustion test of two coals in the bubbling fluidized bed, he also concluded that the SO_2 capture efficiency increased with the temperature.

It was observed from fig. 7(b) that when biomass was mixed with CG, SO_2 emissions increased uniformly with the increase of the combustion temperature. The combustion temperature was increased from 800 °C to 950 °C, SO_2 emissions increased from 340.3 mg/m^3 to 430.6 mg/m^3 when adding WS, SO_2 emissions increased from 324.5 mg/m^3 to 423.6 mg/m^3 when adding CC. Because the sulphur contents in biomass were very small, SO_2 produced by co-combustion was mainly produced by CG. As the combustion temperature increased, the precipitation of inorganic and organic sulphur in CG increased, leading to the increase in SO_2 emissions. Unlike the individual CG combustion, SO_2 release did not show a rapid increase followed by a slow increase as the temperature of the co-combustion samples increased. From the analyses of the test results, it was due to the abundant alkali metal elements in the biomass ash reacted with SO_2 to generate sulphate, which sealed the sulphur in the ash. As the temperature increased, the sulphate was decomposed by heat, causing a small amount of SO_2 to escape.

Comprehensive analyses showed that with the increased of the combustion temperature, both NO and SO_2 emissions increased to varying degrees. The increased in NO was mainly due to the increase of the combustion temperature and the increase in reaction. The increasing rate of SO_2 emission in co-combustion was stronger than that of CG burned alone, because at high temperature, a small amount of sulphate was heated to decompose and generate SO_2.

Conclusions

A laboratory fluidized bed was used to conduct combustion tests on CG and the mixture of biomass and CG. By changing the mixing ratio and the combustion temperature, NO and SO_2 emission characteristics of the samples under different working conditions were studied. Analysing the test results and drawing the conclusions as follows.

- With the increasing of the biomass mixing ratio, NO and SO_2 emissions of the mixed samples decreased. After a large proportion of biomass was added, as the theoretical combustion air volume of biomass was greater than that of CG, under the same air supply volume, a fuel-rich atmosphere was formed in the furnace, resulting in the reduction of NO production. The reduction in SO_2 emissions was due to the reaction of alkali metals and alkaline earth metals, which were abundant in biomass, with SO_2 in the flue gas to form sulphates, which sealed the gas-phase SO_2 in the solid-phase ash, resulting in sulphur fixation.
- With the increase of the combustion temperature, NO and SO_2 emissions from CG burned alone and biomass and CG co-fired both increased. When CG was burned alone, the amount of NO produced increased, and the precipitation of nitrogen-containing compounds in CG became more complete when the temperature rose. The increase in SO_2 emissions was due to the abundance of alkali metal elements in biomass ashes reacting with SO_2 to form sulphates that seal in the sulphur in the ashes. As the temperature increased, the sulphate was decomposed by heat, causing a small amount of SO_2 to escape.

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