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Observation on simultaneously low CO, NO_x and SO_2 emission during oxy-coal combustion in a pressurized fluidized bed

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Abstract: Pressurized oxy-fuel combustion is a promising technology for CO_2 capture offering high combustion efficiency and the simultaneous reduction of gaseous pollutants. A 10 kW_{th} bubbling fluidized bed reactor with continuous coal-feeding was designed, and effects of pressure, temperature and fuel types on pollutant emission were investigated in detail. The relatively low carbon content in the ash and CO concentration in the flue gas demonstrated that the combustion efficiency was improved by high pressure. The concentration of NO, N_2O and SO_2 showed decreased with increasing pressure. Moreover, the effect of pressure on the emission of NO and SO_2 in the lower pressure range (≤0.3 MPa) was more pronounced than that for higher pressure. The concentrations of NO and SO_2 increased with temperature, while N_2O decreased with increasing temperature. Compared with air combustion, NO and SO_2 emission fell sharply in a 21%O_2/79%CO_2 atmosphere. However, N_2O concentration during oxy-combustion was slightly higher than that in air combustion over the range of experimental pressure.

Keywords: pressurized oxy-combustion; fluidized bed; SO_2 emission; NO_x emission.

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1. Introduction

In the foreseeable future, coal will still occupy a significant position in worldwide electricity supply chain[1]. Oxy-fuel combustion is one of the most promising technologies of all the carbon capture and storage (CCS) technologies for coal power plants[2]. A number of facilities of differing scale, up to 35 MW_{th}[3], have been demonstrated[4,5], allowing the exploration of combustion characteristics[6,7], reaction mechanism[8,9] and heat transfer[10,11]. Currently the greatest obstacle to prevent the further development of oxy-fuel combustion is the net efficiency penalty associated with the high energy consumption for the air separation unit (ASU) and compression/purification unit (CPU). Therefore, the critical step for large-scale industrialization is achieving high CO_2 capture efficiency with a minimum energy penalty.

Pressurized oxy-fuel combustion (POFC) technology could be a good solution to this problem. In an atmospheric oxy-combustion system, the ASU and CPU run under elevated pressure, while the boiler runs under atmospheric pressure, so the pressure fluctuations associated with the gas flows cause energy losses and a reduction of net efficiency. When the combustion process occurs under high-pressure conditions, the work losses due to the pressure fluctuations can be substantially reduced. Moreover, it is much easier to recover the latent heat from flue gas in a POFC system. In addition, pressurization avoids the air leakage and reduces the power consumption of the CPU. According to system simulations, the net efficiency of the whole power plant can increase by at least 2% with the combustion pressure of 0.6~1.5 MPa[12–14].

Fluidized bed combustion has its unique advantages in terms of fuel adaptability, especially for low-rank coal, and the combination of fluidized bed and pressurized oxy-fuel combustion has drawn increasing interest in recent years. In addition, pressurized fluidized bed combustion (PFBC) is an established technology in terms of PFBC combined cycle (PFBC-CC) technology for air combustion. To the best of our knowledge, however, only limited

experimental work has been done on POFC due to difficulties encountered in high-pressure operation. Lei et al.[15,16] conducted pressurized coal combustion and emission experiments in a pressurized thermo-gravimetric analyzer (PTGA) and a continuous-feeding fluidized bed, respectively. Their results[15] show the coal ignition mode switches from heterogeneous ignition to homogeneous, and then goes back to heterogeneous as the combustion pressure increases. The PTGA-FTIR (Fourier Transform Infrared) results[16] indicated that 2 MPa is an inflection point for NO_2 and SO_2 emission. When the pressure is lower than 2 MPa, the NO_2 and SO_2 emissions both increase with pressure, while when it is higher than 2 MPa, their concentrations decrease. A pressurized fluidized bed with an inner diameter of 75 mm was used by Lasek et al.[17,18] to investigate the effect of pressure on pollutant emissions and morphologies of ashes from oxy-combustion, and they found that NO, N_2O, and SO_2 emissions were reduced at higher pressures during oxy-combustion, in contrast to the results of Lei et al[15,16]. Lasek`s pioneering research also validated the feasibility of the Flexi-Burn$^{TM}$ technology under elevated pressure, which provided guidance for the future scale-up. However, the maximum pressure of Lasek`s experiment was just 0.44 MPa, which is too low for a POFC system.

Currently, information on pressurized oxy-fuel fluidized bed combustion is still limited, and the mechanisms of how the pressure and atmosphere affect the pollutant emissions have not been clearly explained. A lab-scale pressurized fluidized bed was built for continuous, stable operation. In this work, the effects of pressure, atmosphere and fuel types on gaseous pollutants were investigated to reveal the detailed reaction mechanisms for coal under high pressure.

2. Experimental

2.1 Fuel and bed material

Two different types of fuel, Xiaolongtan lignite and Xuzhou bituminous coal, were used in the experiments. The

fuel was crushed and sieved into small particles with diameters of 0.2~0.35 mm. The sieved particles were heated by an air-blown oven at 120˚C for 3 h to remove the moisture and ensure smooth coal feeding. Table 1 shows the proximate and ultimate analyses of fuel particles after crushing and drying. High-purity quartz sand with a size range of 0.8~1.0 mm was used as the bed material.

Table 1. Proximate and ultimate analyses of fuel (wt, %).

Fuel samples	Ultimate analysis					Proximate analysis			Lower heating value
	$w(C_d)$	$w(H_d)$	$w(O_d)$	$w(N_d)$	$w(S_d)$	$w(A_d)$	$w(V_d)$	$w(FC_d)$	(MJ/kg)
Lignite	53.41	3.36	19.95	1.15	1.71	20.41	43.95	35.64	20.63
Bituminous	70.06	4.43	9.41	0.99	0.59	14.52	31.89	53.59	28.85

*d denotes dry basis; A denotes ash; V denotes volatile; FC denotes fixed carbon.

2.2 Apparatus and procedure

A 10 kW_{th} pressurized fluidized bed system was designed and built, as shown in Figure 1, consisting of a bubbling fluidized bed, electric heater, coal feeder, gas supply lines, temperature and pressure controller and other auxiliary equipment.

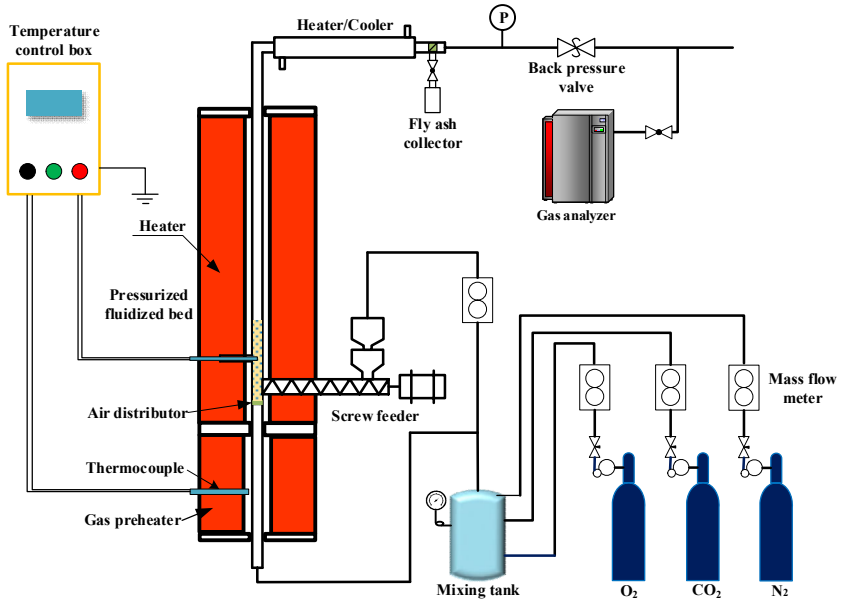


Figure 1. Schematic diagram of 10 kW_{th} pressurized fluidized bed system

The maximum pressure of the fluidized bed was 1.2 MPa with a temperature range of up to 950˚C. The main body of the pressurized bubbling bed consisted of gas preheater, gas distribution plate, combustion chamber and flue gas duct. The primary gas from the mixing tank was heated to 600˚C in the preheater. The combustion section was

made of 253MA stainless steel with an inner diameter of 26 mm and a height of 1250 mm. The gas duct was heated above the acid dew point to avoid condensation. A small high-temperature filter was placed inside the duct to remove and collect large particles (>25 μm) in the flue gas. A backpressure valve was installed at the end of the duct, maintaining stable combustion pressure in the furnace.

Several flue gas sampling ports were included after the backpressure valve to meet the different test requirements. In this experiment, the NOVA Plus gas analyzer (MRU GmbH, Germany) was used to monitor the O_2 concentration in the flue gas, with a resolution of 0.05%, while an Antaris IGS Fourier transform infrared spectroscopy analyzer (FTIR, Thermo Fisher Scientific Inc, USA) was used for the measurement of CO, NO, N_2O and SO_2.

Fuel particles entered the furnace through the screw feeder and pneumatic conveying line. Both the primary and coal-feeding gas came from the mixing tank, and the mass flow controllers (MFCs) were used to adjust the flow rate in the gas lines. The coal-feeding gas usually accounted for 30~35% of the total gas flow. Finally, the gas velocity in the combustor was kept constant at around 0.65 m/s to guarantee good fluidization of the bed material.

During the tests, the bed temperature in the dense zone was set at 800˚C, 850˚C and 900˚C, respectively, and the combustion pressure was controlled at 0.1 MPa, 0.3 MPa, 0.5 MPa and 0.7 MPa to investigate the effect of pressure on the combustion and emission characteristics of the two different fuels.

3. Results and discussion

3.1 Combustion performance

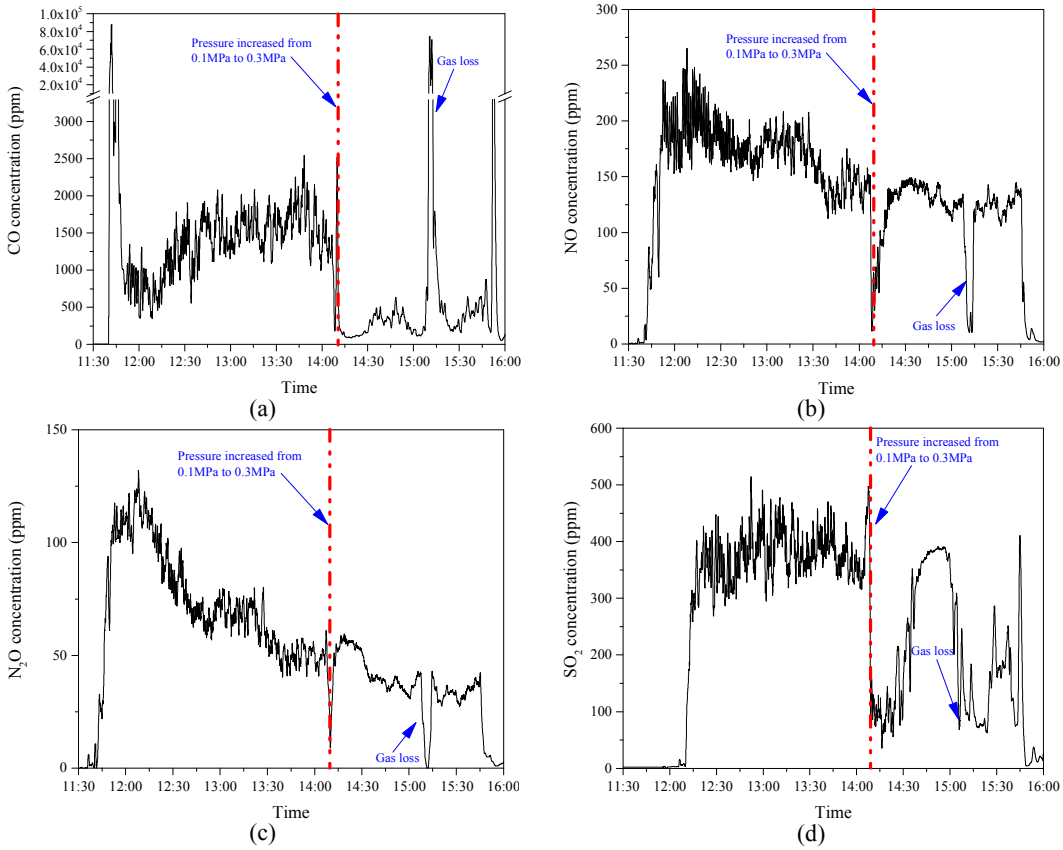


Figure 2. Main exhaust gas compounds (CO, NO_x and SO_2) measured during combustion of lignite in 21%O_2/79%CO_2 at 850˚C.

Figure 2 shows the main exhaust gas compounds measured during combustion of Xiaolongtan lignite at 850˚C in 21%O_2/79%CO_2 atmosphere. The oxygen concentration of flue gas mainly in a range of 6-10%. It is clear from Figure 2(a) that CO experienced a rapid drop from 1856 mg/Nm^3 to 409 mg/Nm^3 when combustion pressure increased to 0.3 MPa. It was found that CO was the most sensitive component to pressure, decreasing significantly even if the combustion pressure was only increased by 0.05 MPa from atmospheric pressure. When combustion pressure of lignite was further increased to 0.7 MPa, as seen in Figure 3, CO dropped to 141 mg/Nm^3, less than 10% of the concentration under atmospheric pressure. The unburnt carbon content in the fly ash was also tested in an elemental analyzer, and results are shown in Table 2.

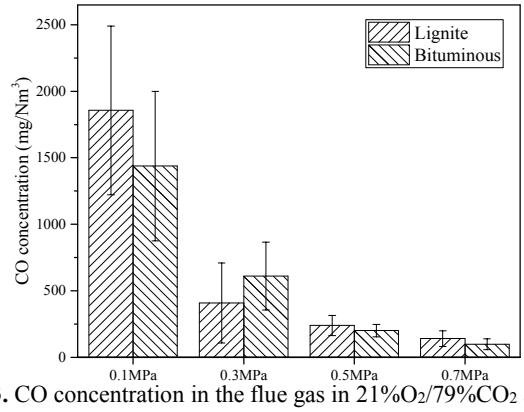


Figure 3. CO concentration in the flue gas in 21%O_2/79%CO_2 at 850˚C.

With the increase of combustion pressure from 0.1 MPa to 0.7 MPa, the unburnt carbon content in the fly ash decreased from 38.5% to 28.7%. Comparing the results to those from the larger-scale oxy-fuel fluidized bed combustor[19,20] and considering the short furnace zone in our experiments, the carbon content after pressurization is relatively low for a lab-scale bubbling bed. The partial pressure of oxygen increases simultaneously with the total pressure, which promotes the oxidation of CO and reduces its emission. Meanwhile, high pressure reduces the bubble size and increase its frequency[21,22], which strengthens gas-solid mixing in the dense phase, and benefits the conversion of carbon. Generally, the low carbon content in fly ash and the reduced CO concentration in the flue gas demonstrated that the combustion efficiency is improved by the high combustion pressure.

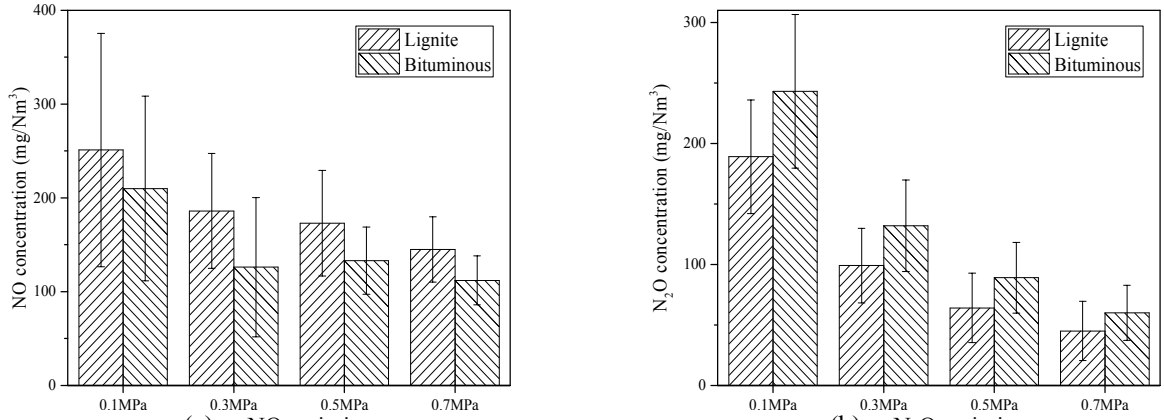
Table 2. Unburnt carbon content in the fly ashes during lignite combustion in 21%O_2/79%CO_2 at 850˚C.

Pressure, MPa	0.1	0.3	0.5	0.7
w(Carbon content), %	38.5	33.1	31.3	28.7

It is also clear from Figure 3 that in most instances when the fuel was changed from lignite to bituminous coal, the CO concentration decreased. The formation and oxidation of CO in the fluidized bed are highly dependent on the combustion conditions and gas mixture. There is no secondary gas in the 10 kW_{th} POFBC, so oxygen decreases along the height of the furnace. Therefore, the char has sufficient time to react with fresh oxygen, while the progressively enhanced reductive atmosphere and the short residence time make reduction of CO in the volatiles more difficult. The CO emissions from bituminous coal at 0.3 MPa and 850˚C are slightly higher than for lignite, which may be explained by the low O_2 concentration (~3.52%) at the furnace outlet.

3.2 *NO_x reactions*

An interesting trend for NO_x emissions was observed during the experiments, shown in Figure 4. Both NO and N_2O decreased with increasing combustion pressure. In the conventional fluidized bed boiler, the behavior of CO is opposite to that of NO_x, and De-NO_x combustion methods needs to create a reducing condition in some specific area of the furnace[23], which leads to an increase in the CO emission. However, pressurized combustion can achieve simultaneous decreases of NO_x and CO, reducing the pollutant emission while ensuring high combustion efficiency.



(a) NO emission (b) N_2O emission

Figure 4. NO_x concentration in the flue gas in 21%O_2/79%CO_2 at 850˚C.

The influence of pressure on NO_x emission can be divided into two aspects: on one hand, high pressure will affect the generation of NO_x in the combustion process; while on the other hand, high pressure may enhance the reduction of NO_x. The pressurized pyrolysis experiments conducted in our previous study[24] found that high pressure promotes the conversion of fuel-N and the yield of NO_x precursors such as HCN and NH_3. The experimental and modeling work on N_2O formation conducted by Mallet et al.[25] also revealed that high pressure promotes the conversion of char-N to N_2O while inhibiting the conversion of volatile-N. What is more, pressure may also effect the interactions of char, CO and NO_x. That is high pressure increases the resistance to NO_x diffusion out of char particles and prolongs the residence time of NO_x in the char pores, which enhances the reduction of NO_x by char.

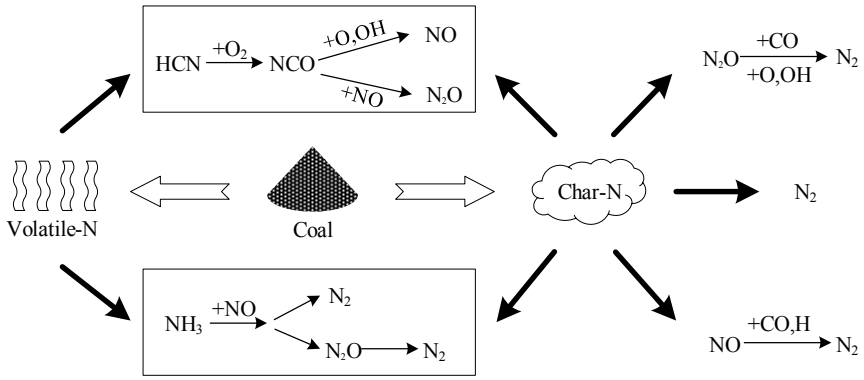


Figure 5. Simplified scheme for the homogeneous reactions of NO and N_2O in a fluidized bed

A simplified scheme for the homogeneous reactions of NO and N_2O is shown in Figure 5. Here, O and OH radicals play an essential role in NO_x formation, and their concentrations may affect the final emission of NO_x. Aho et al.[26] verified this conjecture in a pressurized entrained flow reactor, and their study suggested that the homogeneous formation of NO decreases with increasing pressure. The three-body reaction $H+O_2(+M)=HO_2(+M)$ begins to compete with and finally dominates over the chain branching reaction of $H+O_2=OH+O$ as pressure increases at 850°C. This change will cause the decline of O and OH radicals, as well as the NO concentration.

In general, we can summarize that high pressure: 1) promotes the yield of NO_x precursors like HCN and NH_3; 2) prolongs the residence time of NO_x and enhances the NO_x reduction by char; and 3) reduces the concentration of O and OH radicals and inhibits the formation of NO. Therefore, the effect of pressure on NO_x emission in O_2/CO_2 atmosphere is a combination of fuel-N oxidation and NO_x reduction, and the drop in NO_x concentration under high combustion pressure results from the reduction rate of NO_x being higher than its formation rate.

Figure 4, shows that fuel-N conversion to N_2O is greater in bituminous coal, while the conversion to NO is favored for the low-rank coal. The same trend has been observed in different scale oxy-CFB facilities[27,28]. This could be explained by the different nitrogen components in the coal particles. Pyridines, pyrrole, and quaternary nitrogen are the three principal N-compounds in the coal. A previous study has showed that part of the pyridines and pyrrole nitrogen are converted to HCN during the pyrolysis process[29], which serves as a precursor to N_2O formation. The other NO_x precursor, NH_3, comes mainly from the decomposition of quaternary nitrogen and the secondary reaction

of tar or char. So the different content of specific nitrogen compounds may explain the ambiguity between NO and N_2O formation.

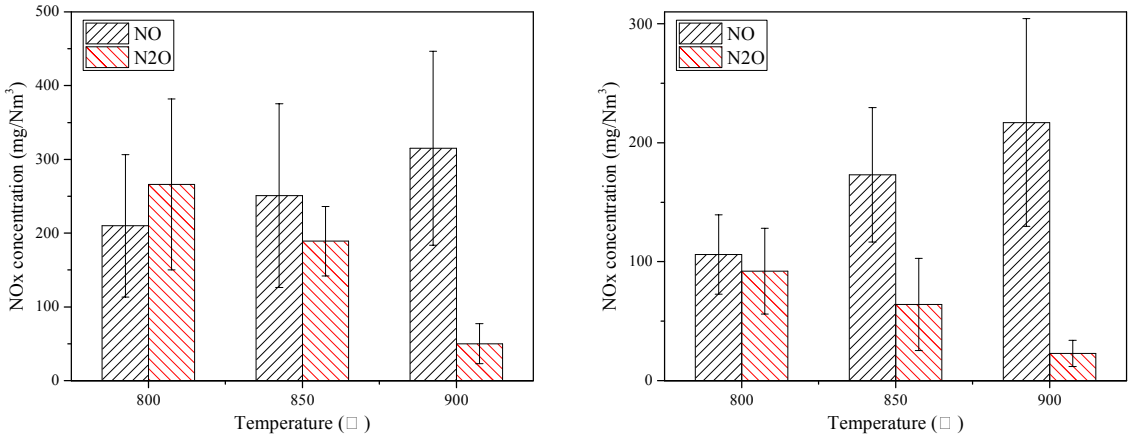


Figure 6. NO_x concentration from lignite combustion at different temperature in 21%O_2/79%CO_2.

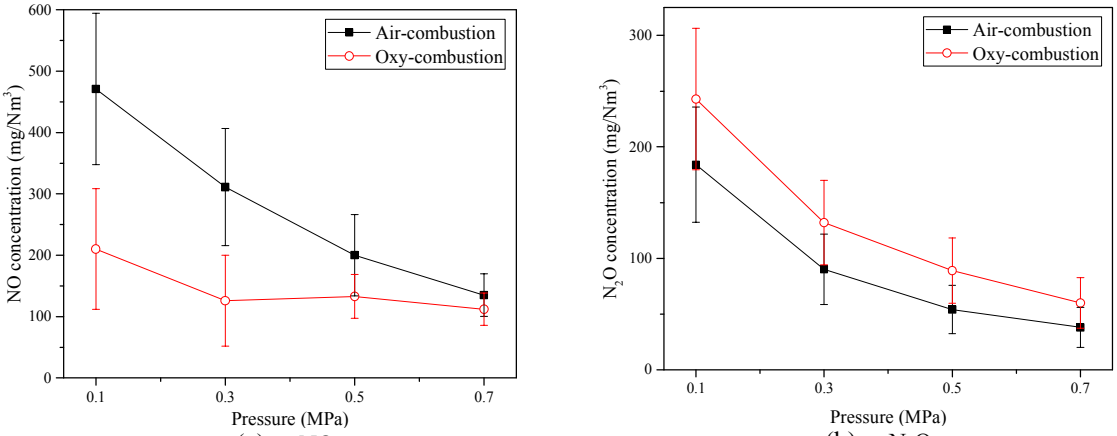
Figure 6 shows the NO_x concentrations from lignite combustion with different temperatures at 0.1 MPa and 0.5 MPa. It is clear that at both pressures, NO emission increases with the bed temperature. The influence of temperature on NO can be divided into several aspects. First, the rise of combustion temperature enhances the conversion of char and the yield of volatile-N. Second, oxygen molecules are more likely to generate O and OH radicals at higher temperature, which makes oxidation of NH_3 and HCN easier. What is more, the lower concentration of CO at high temperature, may slow down the catalytic reaction of NO/CO/char, and increase the final emission of NO.

N_2O emission decreases with combustion temperature in a fluidized bed, and this phenomenon has been widely examined both in air and oxy-combustion. Previous research[30] indicated the following reactions as the main reduction steps of N_2O in the furnace. The rise of temperature significantly accelerates the rate of these reactions. In addition, the formation of O and OH radicals at high temperature also promotes the reduction of N_2O.

$N_2O(+M) = N_2+O(+M)$ R1
$N_2O+O = N_2+O_2$ R2
$N_2O+O = 2NO$ R3
$N_2O+H = N_2+OH$ R4
$N_2O+OH = N_2+HO_2$ R5
$N_2O+CO = N_2+CO_2$ R6

Figure 7 presents the NO_x emission from bituminous coal under different atmospheres. NO and N_2O both have are drastically reduced with increasing pressure in air and O_2/CO_2 atmospheres, similar to the case with lignite, and

the decrease of NO is more pronounced in the lower pressure (≤0.3 MPa) range. NO concentration slightly decreases when the combustion mode is switched from air to oxy-fuel. The main reason for this decrease is that the high CO_2 concentration in the atmosphere promotes the gasification reaction of carbon and is favorable for the formation of CO; thus the high CO concentration further enhances NO/CO/char reduction, resulting in a decrease in NO emission. Unlike NO, there seems to be no convincing explanations for the effects of atmosphere on N_2O emission. Roy et al.[31] observed that N_2O formation was greater under oxy-combustion conditions than in air combustion. By contrast, researchers from ICB-CSIC[31,32] found that N_2O emissions were almost similar under both combustion atmospheres. In our experiments, N_2O is slightly higher in O_2/CO_2 atmosphere and this phenomenon is also observed in the once-through combustion experiment conducted by Hosoda[34]. More detailed research is needed to elucidate the mechanism of N_2O formation and reduction in oxy-fuel atmosphere.



(a) NO (b) N_2O

Figure 7. NO_x concentration of bituminous coal at 850˚C in different atmosphere.

3.3 SO_2 reactions

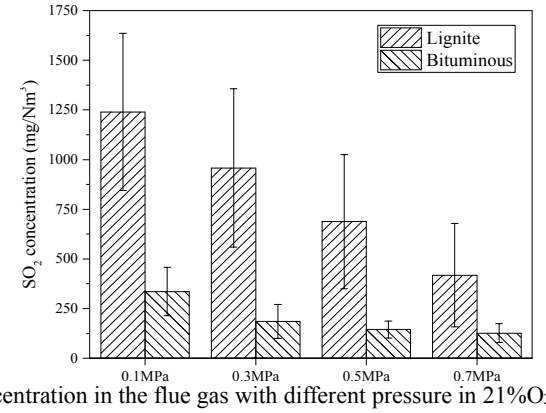
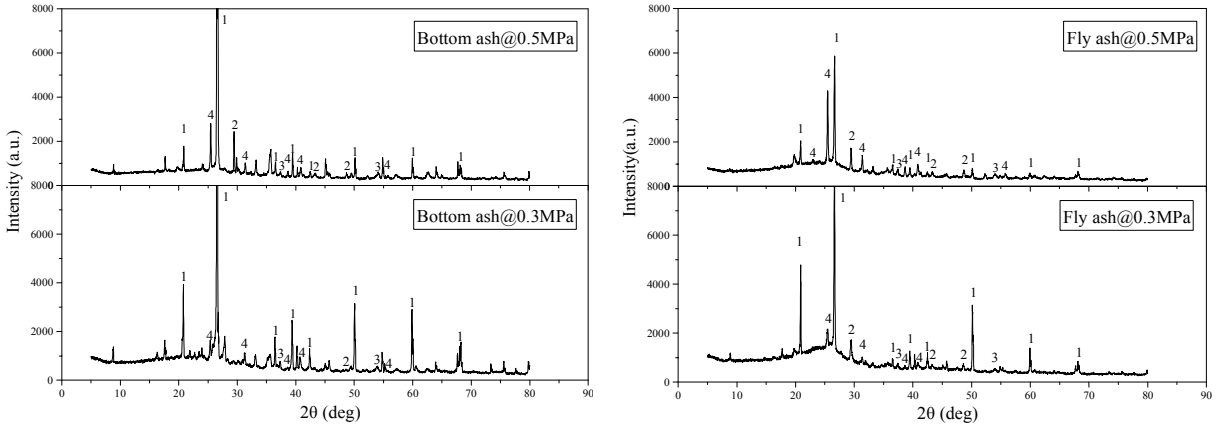


Figure 8. SO_2 concentration in the flue gas with different pressure in 21%O_2/79%CO_2 at 850˚C.

Many parameters, such as atmosphere, temperature, excess oxygen coefficient and fuel type, will affect the SO_2 emission during the combustion process, although the dominant parameter is the sulfur content in the parent fuel. The lower sulfur content in Xuzhou bituminous coal leads to lower emissions of SO_2. SO_2 emissions are also greatly affected by the combustion pressure, as can be seen from Figure 8. SO_2 concentration for lignite decreases from 1240 mg/Nm^3 to 418 mg/Nm^3 as combustion pressure increases from 0.1 MPa to 0.7 MPa, while for bituminous coal, it decreases from 336 mg/Nm^3 to 126 mg/Nm^3.

Previous studies on pressurized fluidized bed combustion found that high pressure could enhance the self-desulfurization ability of ash, and this was verified in our X-ray diffraction (XRD) analysis. Figure 9 shows the XRD profile for fly ash and bottom ash of bituminous coal, here the peaks of SiO_2, CaO, $CaCO_3$ and $CaSO_4$ are marked. It is clear that $CaCO_3$ and $CaSO_4$ are the main components besides SiO_2. The low CaO content proves that the direct sulfation of $CaCO_3$ mainly accounts for the desulfurization mechanism under high CO_2 partial pressure. At the same time, the increase in the relative amount of $CaSO_4$ under higher pressure demonstrates the improvement of the self-desulfurization ability of ash under pressure, which may explain the lower SO_2 emission under higher pressure.



(a)Bottom ash (b)Fly ash

Figure 9. XRD profile for bituminous coal in 21%O_2/79%CO_2 at 850˚C (1-SiO_2;2-$CaCO_3$;3-CaO;4-$CaSO_4$)

Another consideration to explain the decrease of SO_2 may be the SO_3 formation reaction. According to the experiments of Ahn et al.[35], SO_3 emissions were significantly enhanced (4-6 times greater) in an oxy-fuel pulverized-coal test was observed, compared with the air-combustion test. Furthermore, Wang et al.[36] investigated the homogeneous formation mechanism of SO_3 and found that pressurized oxy-fuel combustion shortens the time to reach equilibrium of the overall reaction ($2SO_2+O_2 = 2SO_3$), and promotes the conversion from SO_2 to SO_3.

Figure 10 presents SO_2 concentration from lignite combustion with different temperatures at 0.1MPa and 0.5 MPa during oxy-combustion. The increase of bed temperature leads to a promotion of SO_2 emission. The coal conversion rate rises with the increase of bed temperature and releases more sulfur-containing gases. Moreover, the previous study[37] showed that the sulfur retention capacity of ashes was reduced with the increase in combustion temperature, which also leads to an increase of SO_2 emission.

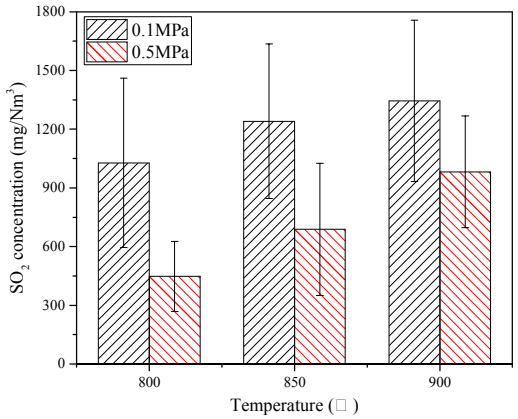


Figure 10. SO_2 concentration from lignite combustion with different temperatures at 0.1 MPa and 0.5 MPa in 21%O_2/79%CO_2.

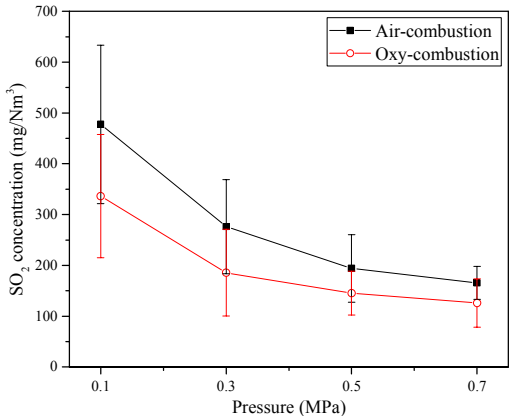


Figure 11. SO_2 concentration of bituminous coal at 850˚C in different atmospheres.

For the oxy-combustion mode, SO_2 concentration is lower than that under air-combustion mode, as shown in

Figure 11. The main forms of sulfur are organic and inorganic, and the main sulfur-containing gas during air-combustion is SO_2. For oxy-combustion, the extremely high CO_2 has a great impact on the conversion of sulfur. A possible explanation is that CO_2, with its high specific heat value, reduces the combustion temperature of the coal particles, enhancing the self-desulfurization ability of ash. Also, the lower CO concentration under high pressure will reduce the possible decomposition of $CaSO_4$ by the reaction:

$$CaSO_4+CO = CaO+SO_2+CO_2 \qquad R7$$

4. Conclusions

Pressurized oxy-combustion experiments conducted in a $10kW_{th}$ fluidized bed allowed us to examine the influences of pressure, temperature, atmosphere and fuel type on pollutant emissions, the following general conclusions can be drawn:

1) Experimental results demonstrate the simultaneous reduction of CO, NO_x and SO_2 in pressurized oxy-combustion, and demonstrate that POFBC offers high combustion efficiency and low emissions.

2) The concentrations of NO and SO_2 increased with temperature, while N_2O concentration decreased. The effect of pressure on the emission of NO and SO_2 over the lower pressure range ($\leq$0.3 MPa) in these experiments was more pronounced than that at higher pressures.

3) Compared with air combustion, NO and SO_2 emissions dropped sharply in 21%O_2/79%CO_2 atmosphere. However, N_2O concentration increased slightly for the oxy-combustion process.

Acknowledgment

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