

Study on NO Emission in the Oxy-Fuel Combustion of Co-Firing Coal and Biomass in a Bubbling Fluidized Bed Combustor

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Combustion experiments were performed in a bubbling bed combustor to explore the effects of various oxygen concentrations (21% to 40%), temperatures (850 °C to 950 °C), and mixing ratios (0% to 30%) on the formation of NO gas. In order to correspond to different combustion stages, the generated NO were distinguished as volatile-NO and coke-NO, respectively, and the total amount and the conversion rate of NO were analyzed. The results indicated that NO comes mainly from fixed carbon combustion, and an increased oxygen concentration in the environment could produce more NO during the combustion process, regardless of the blending of biomass. The temperature increase promoted the conversion of nitrogen in the anthracite and accelerated the release of volatiles from the pine powder. During mixed combustion with increasing temperature, the volatiles formed had diluted the environmental oxygen concentration, which led to suppressed NO emissions. In addition, increasing the blending ratio also resulted in decreased NO emissions due to the large amount of intermediates released by the burning pine powder that induced a deoxidation effect on the NO emission.

Keywords: Anthracite; Oxygen-enriched combustion; Biomass; NO_x emission

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INTRODUCTION

Carbon capture and storage (CCS) technology is an efficient means of mitigating global warming. It is a process in which the wastes of CO₂ are collected at the source of the emissions and transported to a fixed storage site to prevent them from entering the atmosphere. In coal-fired power plants, oxygen-enriched combustion has been widely recognized as being an effective technique for capturing CO₂ and reducing the total CO₂ emissions. The advantages of performing oxy-fuel combustion in a circulating fluidized bed (CFB) boiler includes extensive fuel flexibility, inherently low NO_x emissions, moderate furnace temperature, and effective curing of sulfur by adding limestone, making this the best method for CO₂ capture, and hence emissions reduction. Nonetheless, current technologies are mainly focused on reducing further emissions of CO₂, rather than reducing the concentration of CO₂ already present in the atmosphere. The direct capture of atmospheric CO₂ is a difficult and expensive process (Keith *et al.* 2005).

Fossil fuels will continue to play a major role in energy supply over the next decade. Biomass, as a renewable energy source, has received much attention in controlling the emissions of greenhouse gases. Biomass absorbs CO₂ during its growing process and releases it only when it is burned. It is a fuel that can provide renewable energy as well as temporary storage of CO₂ (Demirbas 2004). The combustion of coal

blended with biomass in a CFB boiler can effectively reduce the emissions of NO_x and SO₂ (Sun *et al.* 2013) gases, and this phenomenon is caused by producing a large amount of reducing gas at the dense phase zone. In addition, the combustion of biomass in an oxygen-enriched environment can effectively inhibit the production of CO₂, make ignition easier and decrease burnout temperatures (Arias *et al.* 2008), and reduce the emission of pollutants (Riaza *et al.* 2012).

Nitrogen oxides are of varying degrees of toxicity and are an important cause of photochemical smog and acid rain (Kampa *et al.* 2008). Over the past 20 years, many researchers have conducted in-depth studies on the conversion of nitrogen and the characteristics of NO_x emissions for various proportions of coal and biomass co-burned in conventional air. Li and Tan (2000), Chang *et al.* (2003), and Mahmoudi *et al.* (2010) are some of the researchers who presented detailed explanations on the NO_x formation mechanism in the pyrolysis stage of coal and biomass. Many researchers have also used their own test designs to study the factors that affect NO_x emissions. Some researchers believe that a temperature rise will promote NO_x emissions (De las Obras-Loscertales *et al.* 2015; Lupiáñez *et al.* 2016; Zhang *et al.* 2016), but others have observed the opposite outcome (Winter *et al.* 1999; Lupiáñez *et al.* 2013) in their experiments. In contrast, many researchers *via* studying the oxygen concentration reached a uniform conclusion that an oxidizing atmosphere promoted NO_x formation (Zhu *et al.* 2015). Regarding the blending ratio, some believed that a combination with biomass could inhibit NO_x emissions (Moroñ and Rybak 2015; Gao *et al.* 2016; Yu *et al.* 2016), while others observed the opposite to be true (Duan *et al.* 2015).

Currently, most studies on NO emissions are focused on final conversion rates and emission concentrations, and few on the phased emissions characteristics. In order to further study the formation rules of NO, volatile-NO and coke-NO (Hayhurst and Lawrence 1996) are considered here with respect to the NO produced in volatiles combustion and fixed carbon combustion stages, respectively. The objective of this study has been to broaden knowledge on this topic through the experimental characterization of NO emissions when two different materials, anthracite and pine, are fired in an oxy-fired bubbling bed reactor.

EXPERIMENTAL

Materials

Instruments and systems

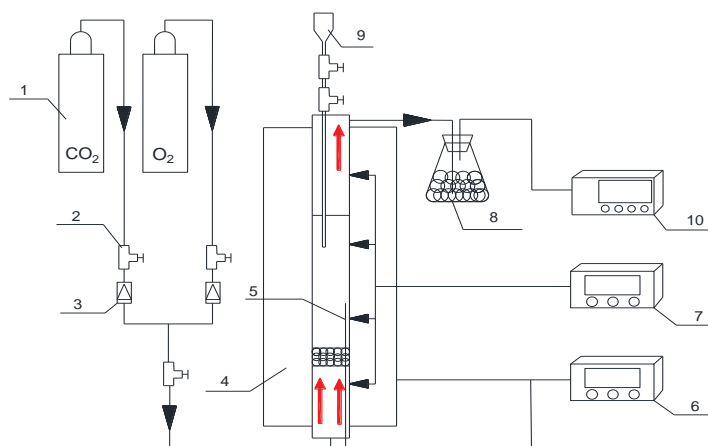


Fig. 1. Schematic diagram of the experimental rig: 1-Gas cylinder, 2-Valve, 3-Mass flow meter, 4-Resistance furnace, 5-K-type thermocouple, 6-Temperature Controller, 7-Pressure Sensor,

8-Anhydrous calcium chloride, 9-Funnel, and 10-Flue gas analyzer

The experimental system used in this chapter is a bubbling fluidized bed combustor. The system structure is shown in Fig. 1. The system consisted of a charging device, electric heating furnace, fluidized bed reactor, a gas supply system, and a flue gas analyzer. High-purity O₂ and CO₂ gases were supplied by gas cylinders. The mixture of gas was adjusted using a flowmeter to the desired ratio in the premix zone. The gas was fed from the bottom of the reaction zone at 2.6 L/min.

The heating device was a vertical tube resistance furnace manufactured by a company in Chongqing (Xinbang Furnace Co., Ltd., Chongqing, China), with a rated temperature of 1000 °C and rated power of 3 kW. The reaction zone consisted of two stainless steel tubes. The outer tube with a diameter of 28 mm and a height of 100 cm was used to isolate the outside air, and it had an internal screen layer that carried the reaction sample. The lower end of the outer tube was connected with the air inlet device, and the upper end was inserted into the inner tube, which was responsible for sending the samples. A screen was placed outside the inner tube near the vent to prevent the sample from blowing out of the reaction zone. The K-type thermocouple was inserted into the furnace at the specified position and the bed reaction temperature was measured; this temperature was then fed back to the temperature controller. A flue gas analyzer (ecom-J2KN, RBR, Iserlohn, Germany) monitored the exhaust gas after it passed through the anhydrous calcium chloride.

Samples of Shandong pine powder (Taian Pingde commerce, Shandong, China) and anthracite (Cangshan coal field, Shandong, China) were used as the biomass and coal samples for the experiment. The raw materials were dried, crushed, ground, and sieved to 300 µm to 500 µm, and the pines were mixed according to pine mass fractions of 0%, 10%, 20%, and 30%, and then stored in sample bags after 2 h for thermostatic drying. The bed material was quartz sand with particle sizes ranging from 300 µm to 600 µm. The data from the proximate analysis and the ultimate analysis of the two samples are shown in Table 1.

Table 1. Proximate Analysis and Ultimate Analysis of Biomass and Coal (%)

Sample	A _{ad}	M _{ad}	V _{ad}	FC _{ad}	C _{ad}	H _{ad}	N _{ad}	S _{ad}
Pine	1.67	12.03	82.16	4.14	46.66	5.62	0.3	0.36
Anthracite	16.19	2.71	8.33	72.77	78.68	2.32	0.8	2.89

*A: Ash, M: Moisture, V: Volatile, FC: Fixed Carbon, and ad: Air Dry Basis.

Methods

Procedures and parameters

The combustion test was first conducted in an electric heating furnace with the bed material (20 g quartz sand) without ventilation. When the reaction interval reached a predetermined temperature, a high concentration of CO₂ was sent through the furnace. The ventilation was stopped after the temperature was stabilized. Then 2 g of the sample was fed from the feeder to the reaction zone, and when the required mixture ratio of O₂/CO₂ was achieved, the measurements were taken.

The concentrations of O₂, CO, CO₂, SO₂, NO, and NO₂ in the exhaust gas were continually monitored in the flue gas analyzer. Many studies have found that the N₂O content in fluidized bed combustion is low and is difficult to measure (Tarelho *et al.* 2011), and hence, this was not included in the flue gas analysis.

Data processing

In this study, the pollutant emission characteristics were analyzed using the following three indexes: 1) the instantaneous concentration of pollutant discharge (ρ), which can be obtained directly through the flue gas analyzer measurements; 2) the total amount of pollutant discharge; and 3) the N conversion ratio, C_N , which represented the percentage of the conversion of nitrogen into NO over the cumulative time period. The C_N showed the precipitation at different stages of the situation, which can be calculated by use of the following formula (Wang *et al.* 2012),

$$C_N = \frac{M_N \cdot V_{gas} \cdot \int_0^t \rho_{NO} dt}{M_{NO} \cdot m_0 \cdot w_N} \quad (1)$$

where C_N is the conversion ratio (%) of N in the sample, and M_N and M_{NO} represent the molar masses of N and NO (g/mol), respectively. The V_{gas} is the flue gas volume flow rate in the standard state (m^3/s). The ρ_{NO} is the concentration of NO in flue gas. The parameter m_0 is the total mass of the sample (mg), w_N is the nitrogen mass fraction in the sample, and t denotes the time (s).

Three types of NO_x emissions are thermal NO_x, prompt NO_x, and fuel NO_x, respectively. The main condition of thermal NO_x formation is the high combustion temperature of up to 1500 °C. A necessary condition for forming prompt NO_x is nitrogen gas. The highest temperature employed in the experiment was not sufficient to generate the thermal NO_x, and the use of CO₂ as the carrier gas instead of N₂ can avoid prompt NO_x formation. So all of the NO_x measured in this experiment was derived from fuel NO_x. To analyze the formation regularity of NO at different stages of combustion, the generated NO was divided into volatile-NO and coke-NO. This corresponded to the NO formed during the volatilization combustion stage and the NO generated in the fixed carbon combustion stage, respectively (Chen *et al.* 2011). All of the full names and abbreviations are listed in Table 2.

Table 2. List of Abbreviations

Abbreviation	Full name
A	Ash
M	Moisture
V	Volatiles
FC	Fixed Carbon
ad	Air Dry Basis
CFB	Circulating fluidized bed
HCN	Hydrogen cyanide

RESULTS AND DISCUSSION

Effect of Oxygen Concentration on NO Emission in O₂/CO₂ Atmosphere

Oxygen-enriched combustion technology has been around for nearly half a century. Also the NO_x formation mechanism has been studied by many researchers (Li and Tan 2000; Chang *et al.* 2003; Mahmoudi *et al.* 2010), and a relatively consistent conclusion has been reached regarding the conventional oxygen-enriched combustion in air. Åmand and Leckner (1991) found that in the conventional air combustion technique, an increased oxygen concentration promoted N₂O and NO emissions, and the effect of the oxygen concentration on N₂O was greater than that on NO emissions. In addition,

Collings *et al.* (1993) reported that the effect of increased oxygen concentration was stronger at lower temperatures than at higher temperatures. These conclusions were subsequently confirmed in various other studies.

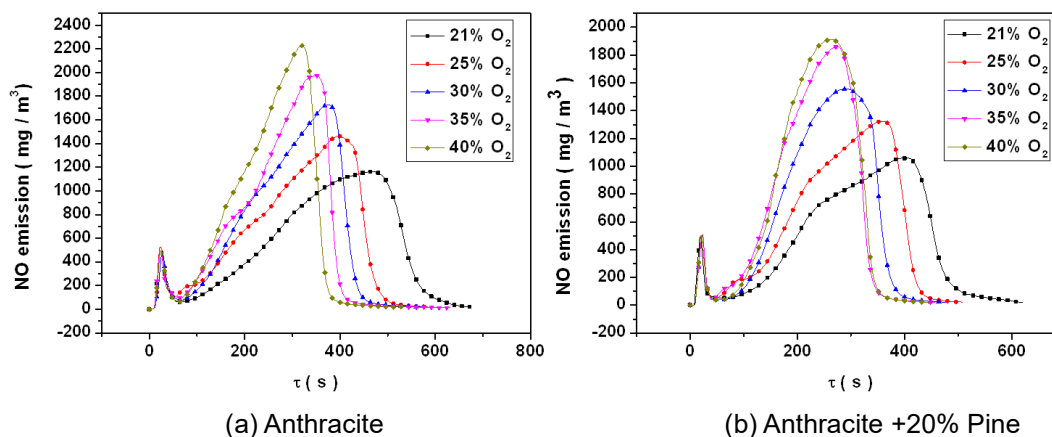


Fig. 2. Instantaneous values of NO emission at 900 °C

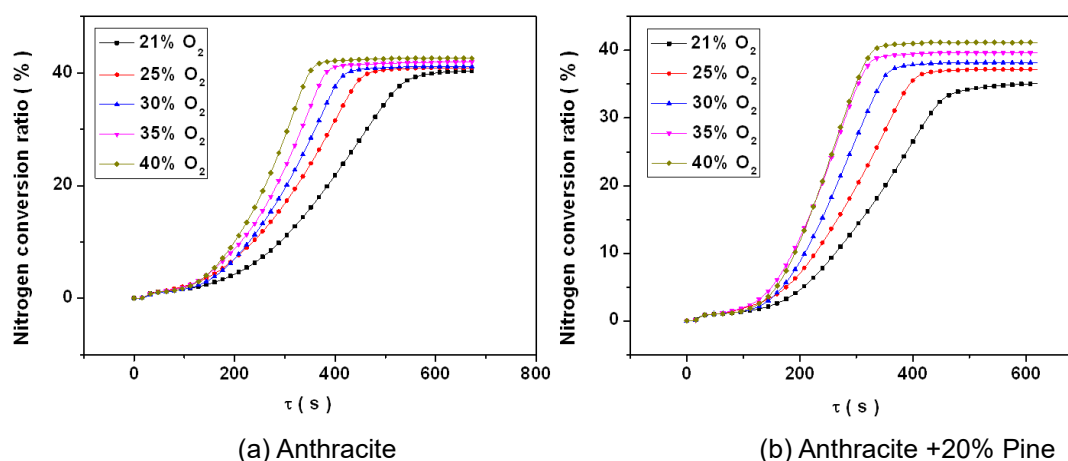


Fig. 3. Instantaneous values of nitrogen conversion ratio at 900 °C

Figure 2 shows the instantaneous values of NO emission with various oxygen concentration levels at 900 °C. The two experimental samples (coal, and coal mixed with 20% biomass) showed similar changes. It was clear from Fig. 2 that there were two distinct peaks in the NO emission curves. In the volatile combustion stage, the NO emission curves with various oxygen concentrations almost coincided with each other, with all of the five peaks at approximately 510 mg/m³. This result revealed that the oxygen concentration level has little effect on the NO emission in the volatile combustion stage.

In the fixed carbon combustion stage, the emission peak value of anthracite (a) increased from 1162 mg/m³ to 2226 mg/m³ with an increase in the oxygen concentration, and the peak value of coal mixed with 20% biomass (b) also increased from 1058 mg/m³ to 1911 mg/m³. The increased oxygen concentration improved the combustion rate, reduced the burnout time, and accelerated the generation of NO, which made the two peaks close to each other.

Figure 3 shows the instantaneous values of the nitrogen conversion ratio with various oxygen concentration levels at 900 °C. It is evident from this figure that the increased oxygen concentration led to an increased rate of NO formation and an increase in the overall conversion rate.

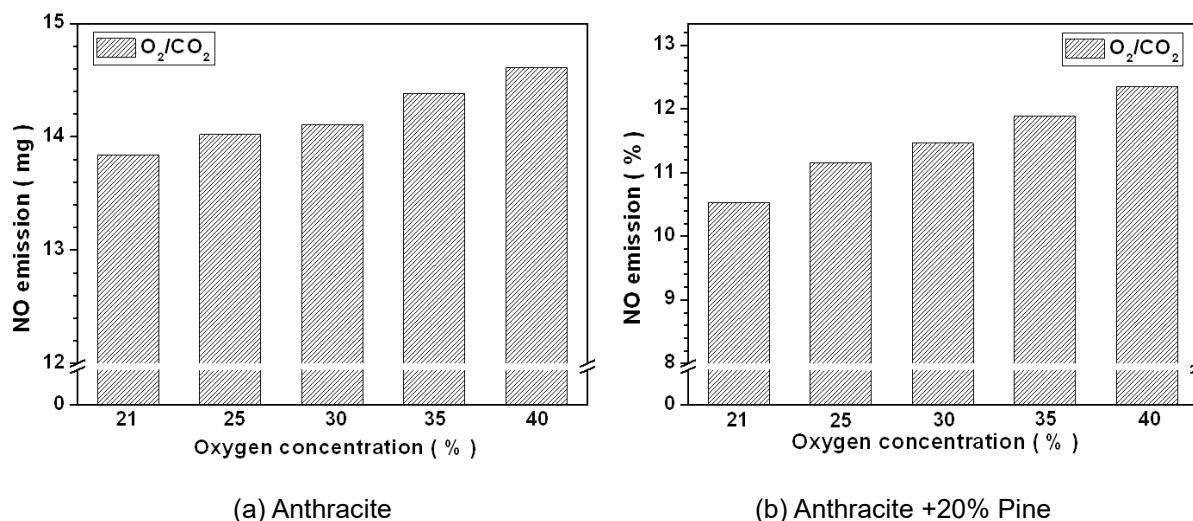


Fig. 4. Total amount of NO emission at 900 °C

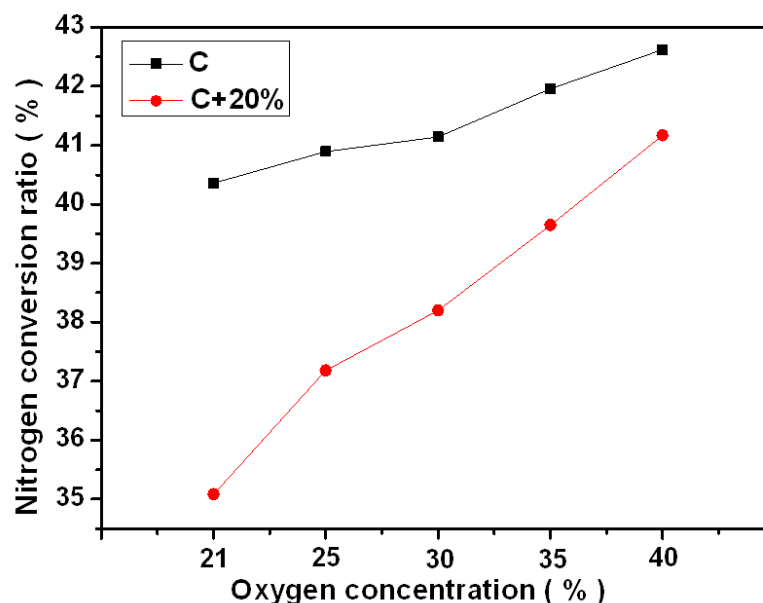


Fig. 5. Conversion rate of fuel-N to NO for combustion at 900 °C

Figure 4 shows the total NO emissions of anthracite and anthracite mixed with 20% pine powder under various oxygen concentration levels. With increased oxygen concentration, the NO emission of anthracite increased slightly from 13.84 mg to 14.61 mg, and that of coal mixed with 20% biomass increased slightly from 10.52 mg to 12.35 mg. Figure 5 demonstrates the conversion rate of fuel-N to NO under various experimental conditions. With increased oxygen concentration, the nitrogenous conversion rate of anthracite increased slightly by 2.27%, from 40.35% to 42.62%. In comparison, the nitrogenous conversion rate increased by 6.09%, from 35.08% to 41.17%, after blending with 20% pine powder. This suggested that the oxygen concentration had a greater effect on the sample mixed with pine.

There are several reasons (Chen *et al.* 2011) that explain why a higher oxygen concentration promoted the NO_x emission. For one, the NO conversion could be divided into three stages: 1) volatile decomposition; 2) volatile pyrolysis product intermediates, such as hydrogen cyanide (HCN) and NH₃, which were further oxidized to NO; and 3) coal char nitrogen that was converted to HCN, which then generated the NO. The possible reactions between the intermediate products and NO are described as follows:



As the partial pressure of oxygen increased in the atmosphere, the adequate amount of oxygen present would promote the above two reactions, and this resulted in an increased total amount of NO emissions. The char burned more completely, and less CO, H₂, and hydrocarbons would be produced in high oxygen concentration levels, thus a reduced chance of decreases in char/NO/CO (Duan *et al.* 2015). The char, CO, H₂, and hydrocarbons also burned more completely in the high oxygen concentration, thus a reduced chance of NO deoxidation. In addition, the total gas flow rate became lower, and the residence time of the gas in the furnace became longer at higher oxygen concentrations. Therefore, more fuel combustions and less NO reductions were also possible and caused the slight increase of NO conversion with increased oxygen concentrations (Zhu *et al.* 2015).

Effect of Combustion Temperature on NO Emission in O₂/CO₂ Atmosphere

Figure 6 shows the total NO emissions of the two fuels at varying combustion temperatures in a 35% O₂/65% CO₂ atmosphere. With an increased combustion temperature, the total NO emission of the anthracite increased slightly from 14.04 mg to 14.72 mg. In comparison, the total NO emission was reduced from 12.48 mg to 11.46 mg after blending with 20% pine powder. The conversion curve of fuel-N to NO in Fig. 7 highlights the effect of the combustion temperature on the conversion rate.

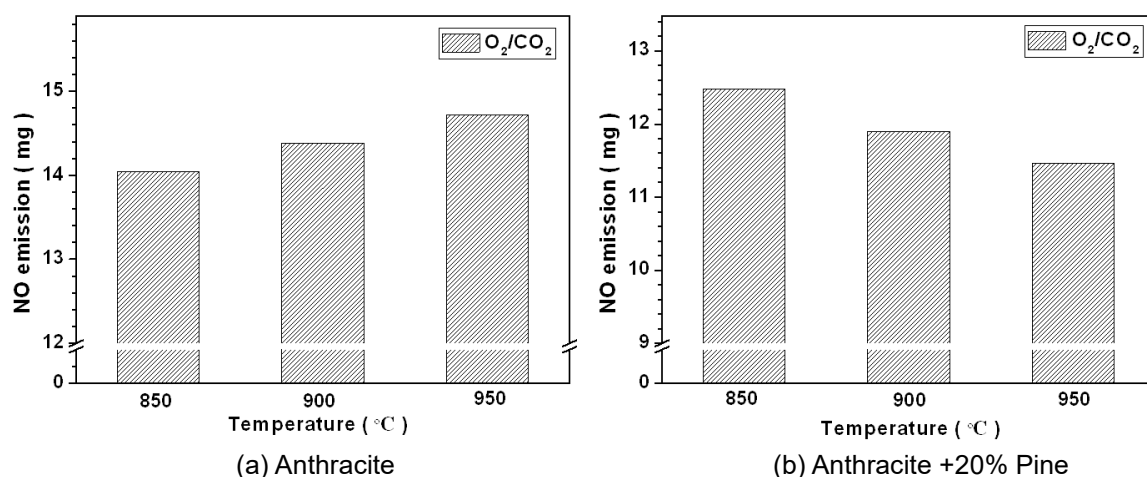


Fig. 6. Total values of NO emissions (35% O₂ /65% CO₂ atmosphere)

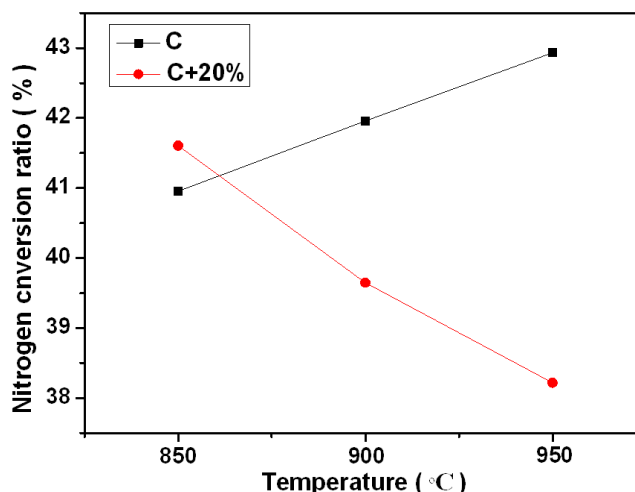


Fig. 7. Conversion of fuel-N to NO_x for combustion at 35% oxygen concentration

From 850 to 950 °C, the fuel-N conversion rate of anthracite increased by 1.98%, but the fuel-N conversion rate of coal mixed with 20% pine powder decreased by 3.39%. Other researchers have also observed a reduction in the NO emissions with an increased combustion temperature (Winter *et al.* 1999; Gou *et al.* 2007; Bai *et al.* 2013; Zhu *et al.* 2015). In fact, the increase in temperature accelerated the release of nitrogen compounds in the fuel and promoted the conversion of NO, which increased the conversion of fuel nitrogen and the concentration of NO in flue gas. When the blending amount reached above 20% as the temperature rose, the release rate of the volatile matters in the fuel became fast enough that the residence time of the gas in the reactor became shorter. This would weaken the oxidization process of the nitrogen-containing intermediate products to form NO. In addition, given that the oxygen diffusion in CO₂ was slower (Lupiáñez *et al.* 2013), the reduction of NO over char particles in the bed must be kinetically increased by elevating the temperature, because the kinetic increase was possibly much larger than the decrease, owing to the shortened gas residence time at elevated temperatures (Jones *et al.* 1999). For these reasons, with coal mixed with 20% pine powder, the NO emission trends in Figs. 6 and 7 decreased with the increase in the combustion temperature. Almost no other coal combustion experiments have shown this result (Gou *et al.* 2007).

It is worth noting that in Fig. 7 at 850 °C, the nitrogen conversion percentage of the mixed fuel was 41.60%, which was higher than that of the anthracite conversion rate of 40.95%. This means that the reduced amount resulted from the low nitrogen content of the pine powder. When the temperature reached 900 °C and above, the nitrogen conversion rate of the mixed fuel was lower than that of the anthracite, which indicated the presence of a synergetic effect after the pine powder mixing.

Effect of the Biomass Blending Ratio on NO Emission in O₂/CO₂ Atmosphere

Figure 8 shows the instantaneous rates of change in the NO emissions and their corresponding fuel-N conversion rates with different blending ratios at 950 °C in a 35% O₂/65% CO₂ environment. Figure 8(a) clearly shows that the pine powder had a weak effect on the NO emission in the volatile combustion stage but had a substantial effect in the fixed carbon combustion stage. Its emission peak decreased from 1857 mg/m³ to 1312 mg/m³. Figure 8(b) clearly indicates that with the increase of the mixing ratio, the NO release rate was correspondingly accelerated, but the fuel nitrogen conversion rate gradually decreased.

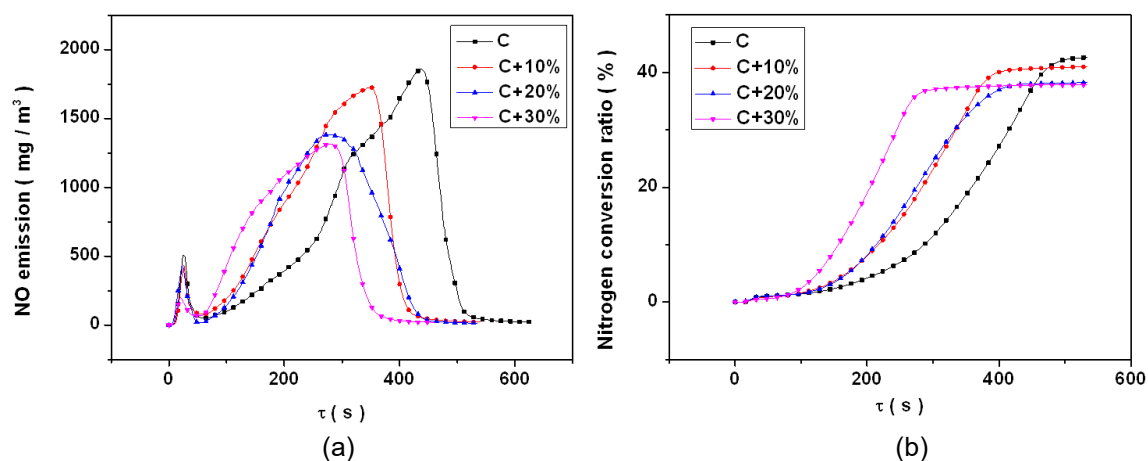


Fig. 8. The NO emission and nitrogen conversion ratio curves of anthracite and anthracite mixtures of different proportions

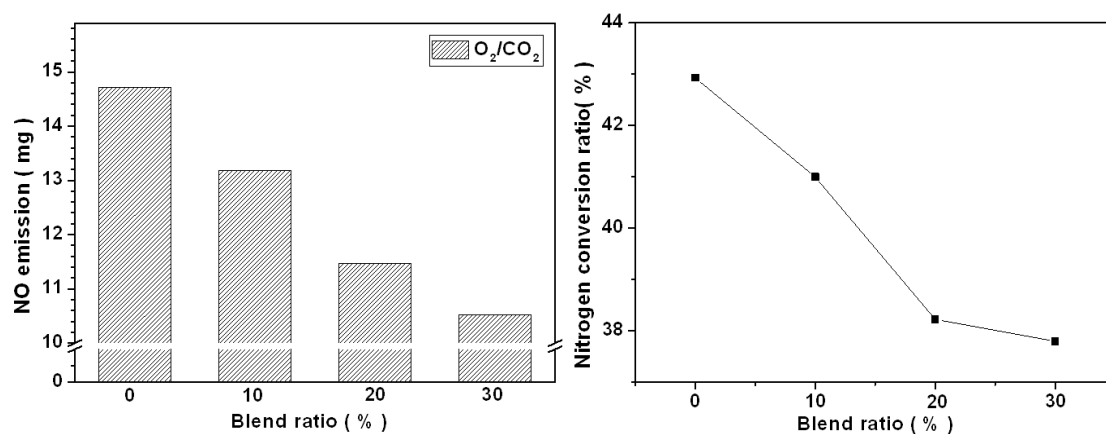


Fig. 9. The NO emission and nitrogen conversion ratio at different biomass mixing ratios

Figure 9 shows the changes in the total NO emissions and their corresponding total fuel-N conversion rates with different blending ratios at 950 °C in a 35% O₂/65% CO₂ environment. It was clear that the mixed pine powder had different inhibition effects on the total NO emission and the total fuel nitrogen conversion. The emission of NO was reduced from 14.72 mg to 10.53 mg. Similarly, the conversion of fuel nitrogen was decreased from 42.93% to 37.79%. The inhibitory effect was enhanced with an increased blending ratio.

This phenomenon could be explained as follows: At first, because the nitrogen content in biomass was lower than the nitrogen content in anthracite, blending biomass could reduce the total nitrogen content of the fuel. Using this blended fuel could reduce the level of NO emission from the combustion. Secondly, the biomass pyrolysis would gradually release a large number of nitrogen-containing intermediates. According to a study by Glarborg *et al.* (2003), regardless of the degree of carbonation of the fuel, the N elements in the fuel were mainly released in the form of HCN and NH₃. In the process of NH₃ being oxidized to NO, the formed amino radicals, such as NH and NH₂ are important intermediates. The NH and NH₂ radicals could react with NO to produce N₂ or N₂O (Reactions 4 through 8) (Miller and Bowman 1989; Johnsson 1994; Okazaki and Ando 1997). The produced HCN also played an important role in the reduction of NO to N₂O. The most important reactions were, (9) and (10), as shown below (Johnsson 1994; Glarborg *et al.* 2003):



Furthermore, with an increased amount of pine powder, a high level of volatiles that were produced would dilute the oxygen concentration, which caused more coke to react under high concentrations of CO_2 in the environment and produce a large amount of CO (Duan *et al.* 2011). Although the intermediate products containing nitrogen were more easily oxidized by oxidizing gases, than nitrogen in coke, they were also consumed by reducing gases such as hydrocarbons, carbon monoxide (CO), and other reducing gases (Hayhurst and Lawrence 1996). The reaction equation is as follows,



In addition, according to Li and Tan (2000; 2003), a high concentration of CO_2 could suppress the formations of HCN and NH_3 by consuming the N-sites or H radicals on the char surface, where HCN and NH_3 were the main NO precursors during the combustion process.

CONCLUSIONS

An emission characterization of anthracite and pine powder co-firing under the condition of O_2/CO_2 has been studied in a lab-scale bubbling fluidized bed. CO_2 instead of N_2 as the carrier gas can effectively eliminate the thermally induced NO and prompt the generation of NO . Compared with the volatile combustion stage, the fuel- NO mainly comes from the fixed carbon combustion stage. Whether or not the biomass is blended, an increase of oxygen concentration may improve combustion, but will produce more NO in the combustion process.

Due to the high content of volatiles in the biomass, when the blending amount of pine powder was 20% or above, the temperature rise suppressed the generation of NO . When the reaction temperature was above $900\text{ }^\circ\text{C}$, the fuel nitrogen conversion rate of the mixed pine powder became lower than that of the anthracite. This indicated that the mixed pine powder had a synergetic effect.

Due to the fact that the nitrogen content of pine powder was lower than that of a mixture of anthracite and pine powder, a large number of intermediate products were released during combustion. Because of this, coal blending with biomass can effectively inhibit the generation of fuel- NO . Moreover with the increase of the blending amount, the inhibition effect became obvious.

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