


RESEARCH ARTICLE

NO_x emissions of pulverized coal combustion in high-temperature flue gas

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Abstract

An improved high-temperature flue gas combustion technology was developed and examined in this study. The high-temperature flue gas generator generated a flow of flue gas at a high temperature (720°C) with low oxygen content (10%), which flowed into the main combustion zone and acted as the carrier gas for pulverized bituminous coal (0–0.355 mm) in the combustion chamber. In the combustion chamber, the coal particles reached a higher combustion temperature (>1,100°C) in a short time, and the temperature difference was within 230°C, which showed that the local oxidation zone had been avoided. Furthermore, air-staging combustion technology was also adopted in combination with this technology. NO was the main nitrogen-containing compound, and the concentrations of other nitrogen-containing compounds (N₂O, NH₃, and HCN) were extremely low. The NO concentrations were reduced to 0 ppm twice when the volatiles released rapidly in the initial stage, and the volatiles burned violently in the intermediate stage, which are the main reasons for the significant reduction in NO_x emissions. The original emissions of NO_x were 187.2 mg/m³ (@6% O₂), and the conversion ratio of fuel-nitrogen to NO_x was 16.6%. High-temperature flue gas combustion technology has reached low-NO_x combustion for pulverized coal.

KEYWORDS

air-staging, coal combustion, high-temperature flue gas, NO_x emissions

1 | INTRODUCTION

Combustion is one of the main means of utilization in coal energy.^{1,2} Obtaining high combustion efficiency and low pollutant emissions has always been a hot topic in the field of coal combustion research.^{3,4} On this hot topic, reducing nitrogen oxide (NO_x) emissions has received the most attention as a difficult part of controlling pollutant emissions.^{5,6} In the conventional combustion mode, the high combustion efficiency of pulverized coal is usually attained by increasing the combustion temperature, but the amount of thermal NO increases exponentially when the combustion temperature

exceeds 1,500°C,^{7,8} leading to a dramatic increase in NO_x emissions. Technologies that attain low-NO_x emissions during conventional combustion processes include low-NO_x burner technology^{9,10} and staged combustion technology.^{11,12} Staged combustion technology includes air staging and fuel staging, which refers to the air or fuel being injected into the combustion chamber (CC) in a layered manner, thereby establishing several reductive zones in the CC and increasing the reduction ratio of fuel-nitrogen. It should be noted that an unreasonable reductive zone would cause a reduction in combustion efficiency.^{13,14} In summary, achieving clean and efficient coal combustion requires a balance among the

variations in the two indicators of high combustion efficiency and low-NO_x emissions.

The high-temperature air combustion (HTAC) of pulverized coal is a promising clean and efficient combustion technology. HTAC technology was first applied in gaseous fuel combustion. Suzukawa et al.,¹⁵ Fujimori et al.,¹⁶ and Katsuki and Hasegawa¹⁷ devised an alternating flow of air and flue gas in a ceramic honeycomb. The stored thermal energy of the flue gas in the ceramic honeycomb was used to heat the air to attain HTAC for gaseous fuel. The results show that NO_x emissions were drastically reduced. This technology has advantages like high thermal conductivity and low noise, based on which some scholars have applied this technology to solid fuel combustion. However, the heating mode of alternating flow in the ceramic honeycomb cannot be applied in the pulverized coal combustion process because the dust-containing flue gas would block the gas circulation pipeline in the honeycomb. Suda et al.¹⁸ and Zhang et al.¹⁹ reached HTAC in pulverized coal by adopting electric air-heating furnaces or primary air enrichment and preheating (PRP) burners, respectively, where the PRP burner introduced the flue gas into the burner chamber and the flue gas mixed with air in advance to raise the air temperature. The results show that the burnout efficiency of pulverized coal was improved and NO_x emissions were drastically reduced. The immense potential of HTAC in pulverized coal combustion has been verified.

The mechanism of low-NO_x emissions in the HTAC of pulverized coal¹⁸ is that the combustion reaction rate of high-temperature air with pulverized coal accelerates at the entrance to the CC, which greatly increases the devolatilization rate of the pulverized coal, creating a local reducing atmosphere and promoting the irreversible reduction of fuel-nitrogen to N₂ during the initial release period. Furthermore, the reduction ratio in this zone could be further increased by gas dilution, that is, a dilution of pulverized coal carrying gas would reduce NO_x emissions. Saha et al.^{20–23} indicated that coal and biomass could both realize an efficient low NO_x in a furnace supplied with a hot and vitiated coflow, which is supplied by a secondary burner with a turbulent nonpremixed swirl flame of natural gas. Moreover, Weidmann et al.^{24,25} summarized that diluting the reactants resulted in a low local heat release and low temperature increase in the reaction zone. Because a relatively low-temperature reaction environment could reduce the NO_x generated from fuel-nitrogen combustion at a specific reaction rate, it is beneficial for realizing low-NO_x emissions. In addition, numerous numerical studies have proved that diluting the reactants contributes to reducing the combustion peak temperature and NO emissions.^{26,27} It should be noted that it is easier to reach low-NO_x emissions by

diluting the carrier gas than by diluting the combustion oxidant (air).²⁵

In previous studies,^{28,29} we had conducted related experiments on diluting the combustion oxidant: high-temperature flue gas was generated by a precirculating fluidized bed (CFB), and the high-temperature flue gas from the CFB was used as the primary air for coal combustion in a rear CC. The results show that, compared with conventional combustion technologies, NO_x emissions could be drastically reduced while maintaining high combustion efficiency. However, the fuel consumption in the CFB accounted for a sizable proportion of the total fuel consumption in the previous study. Therefore, the combustion share in the main CC was reduced, which resulted in heat waste, and the results could not provide theoretical guidance for the capacity enlargement of the equipment. Therefore, it is necessary to reduce the size of the CFB. In addition, the coal feeding rate to the CFB needs to be drastically reduced, so the main combustion share moves into the CC. Moreover, the high-temperature flue gas generated by the CFB was adopted as the carrier gas for the pulverized coal, which could further improve the reduction degree of NO_x.

In this study, air-staging combustion technology was adopted in combination with the improved high-temperature flue gas combustion technology to reduce NO_x emissions. Furthermore, we describe the migration and transformation path of fuel-nitrogen in the main combustion zone in detail to obtain new control measures for low-NO_x emissions from pulverized coal combustion. The results could provide a theoretical basis for future engineering practices.

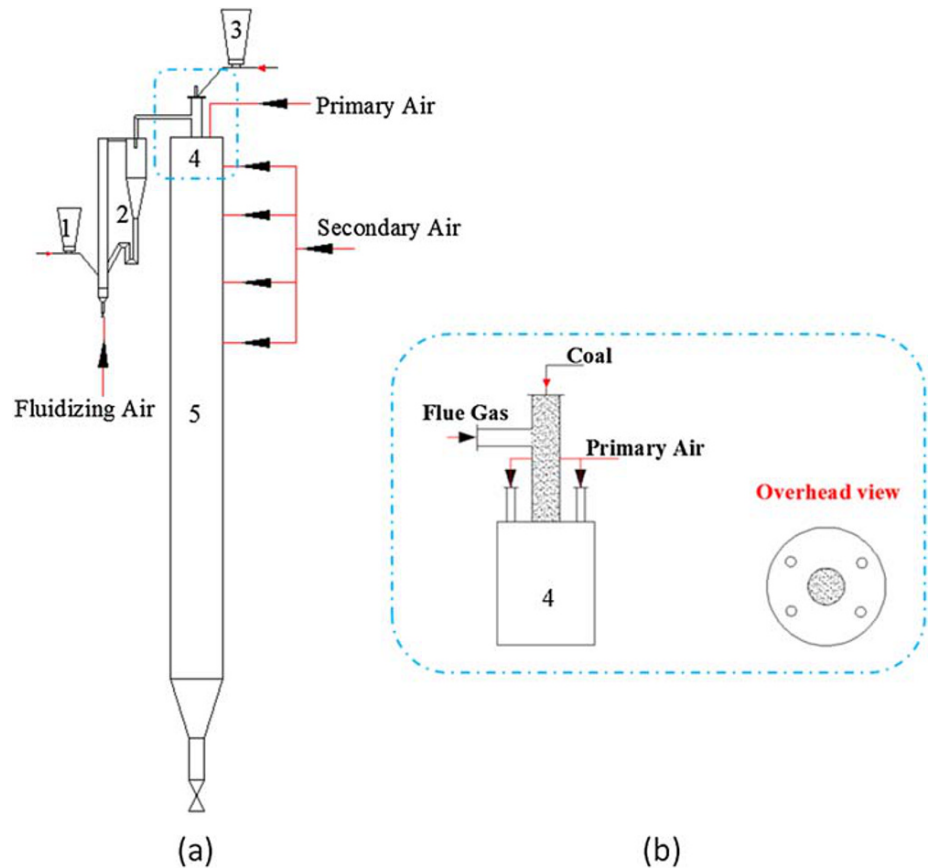
2 | EXPERIMENTAL

2.1 | Test system and method

The test system was mainly composed of a high-temperature flue gas generator (HTFG) and a CC. A schematic diagram of the test platform is shown in Figure 1a. The test process can be briefly described as follows: A small amount of pulverized coal reacted with the fluidizing air and underwent a complete combustion reaction, which produced a high-temperature flue gas in the HTFG. Then, the high-temperature flue gas carried the experimental coal into the CC, and the combustion reactions proceeded from top to bottom.

The structure of the HTFG was a CFB made of Cr25Ni20 steel. The height and diameter of the riser were 1,000 and 149 mm, respectively. Three thermocouples (Ni–Cr/Ni–Si) were arranged at 50, 500, and 950 mm above the bottom of the riser, and two other

FIGURE 1 (a) Schematic diagram of test platform (1. Coal feeder; 2. High-temperature flue gas generator; 3. Coal feeder; 4. Combustion burner; 5. Combustion chamber). (b) Burner nozzle structure at the CC top



thermocouples (Ni–Cr/Ni–Si) were arranged at the returner and outlet of the cyclone. In addition, three pressure-measuring points were arranged at 50 and 950 mm above the bottom of the riser as well as the outlet of the cyclone to monitor the pressure drop of the riser and the cyclone.

The structure of the CC was cylindrical, with a height and diameter of 6,000 and 700 mm, respectively. The CC was mainly composed of refractory materials. In addition, there were 17 thermocouples (Pt/Pt–Rh) arranged every 300 mm for 150–3,150 mm downward and every 500 mm for 3,500–6,000 mm downward below the CC top. The measurement error of the combustion temperature was within $\pm 5^\circ\text{C}$.

The burner nozzle structure at the CC top is shown in Figure 1b. The high-temperature flue gas flowed into the main nozzle pipe from the left-side pipe, and the pulverized coal was added to the main nozzle pipe from the upper part of this pipe. At the intersection of these two mass flows, the high-temperature flue gas carried pulverized coal from the top central main nozzle pipe, which had a diameter of 51 mm, to the CC. The distance between the intersection and the exit of the burner nozzle was approximately 760 mm, and the gas velocity in this pipe was approximately 12–14 m/s, so the reaction time between the high-temperature flue gas and coal in

the transport pipe was short. In addition, the oxygen concentration in the high-temperature flue gas was low, so the reaction rate was low, which ensures mixing and flow without slagging and blocking in the transport pipe. The primary air flowed into the CC from four annularly distributed gas pipes, each of which had a diameter of 30 mm. The secondary air flowed into four air ducts of the same size (diameter: 28 mm) from the main air duct, and these four air ducts were arranged at 300, 1,200, 2,400, and 3,000 mm below the CC top. In addition, the temperatures of the primary air and the secondary air were both 20°C , and thermal mass flow meters were adopted to obtain exact flow rates.

A high-temperature flue gas sampling point was arranged at the cyclone outlet. There were nine flue gas sampling points arranged every 600 mm for 150–5,500 mm below the CC top. The tail flue gas from the test system was sampled from the pipe of the flue gas water cooling outlet, and the component was also defined as the flue gas component at 6,000 mm below the top. The flue gas components (NO , N_2O , NH_3 , HCN , CO , and CO_2) were measured using a Gasetm FTIR DX-4000 analyzer, and the oxygen concentration in the tail flue gas was measured using a zirconia oxygen analyzer. The measurement error of the gas concentration was within $\pm 2\%$.

2.2 | Experimental condition

The experimental fuel was bituminous coal with a particle size range of 0–0.355 mm. The proximate and ultimate analyses are summarized in Table 1.

The global air equivalent ratio (λ_{Stoic}) is calculated based on the oxygen concentration in the tail flue gas, which is the oxygen concentration after the complete combustion of the total coal (including coal feeding rates in HTFG and CC) and total air (including the fluidizing air, primary air, and secondary air) in the whole system. The definitions of the air equivalent ratios of the fluidizing air (λ_{FA}), primary air (λ_{PA}), and secondary air (λ_{SA}) are summarized as follows:

$$\lambda_{\text{FA}} = \frac{A_{\text{FA}}}{A_{\text{Stoic}}}, \quad (1)$$

$$\lambda_{\text{PA}} = \frac{A_{\text{PA}}}{A_{\text{Stoic}}}, \quad (2)$$

$$\lambda_{\text{SA}} = \frac{A_{\text{SA}}}{A_{\text{Stoic}}}, \quad (3)$$

$$\lambda_{\text{Stoic}} = \frac{21}{21 - O_{\text{FG}}}, \quad (4)$$

$$A_{\text{Stoic}} = (A_{\text{FA}} + A_{\text{PA}} + A_{\text{SA}}) \times \lambda_{\text{Stoic}}, \quad (5)$$

where A_{FA} , A_{PA} , and A_{SA} (Nm^3/h) are the fluidizing air flow rate in HTFG, the primary air flow rate, and the secondary air flow rate in CC, respectively. A_{Stoic} (Nm^3/h) is the air flow rate for stoichiometric complete combustion, and O_{FG} (%) is the oxygen concentration in the tail flue gas.

During stable operation, the input thermal power of the test system was 0.113 MW. The ratio of coal feeding rate in the HTFG to the CC was approximately 1:9. In addition, the jet Reynolds numbers of the primary air and secondary air were 2,622 and 6,367, respectively. The time average value of the oxygen concentration in the tail flue gas was 3.93%, so the global air equivalent ratio of the test system was 1.23. Detailed experimental operating parameters are summarized in Table 2.

3 | RESULTS AND DISCUSSION

3.1 | High-temperature flue gas generator

The temperature variation and pressure drop variations with time when the coal feeding rate and the fluidizing air flow rate in the HTFG were 1.62 kg/h and 25.8 Nm^3/h , respectively, are shown in Figures 2 and 3. Here, the temperature refers to the center temperature of the plane. Within the same time period (180 min), the temperature and pressure drop both fluctuated steadily, and the variation in nearly all combustion temperatures was within $\pm 3\%$, which indicates the operating stability of the HTFG. Figure 2 indicates that the time average value of the four main combustion temperatures in the HTFG was approximately 920°C, and the time average value of the flue gas temperature at the cyclone outlet was approximately 720°C, that is, the HTFG could stably generate a flow of high-temperature flue gas at 720°C.

The high-temperature flue gas components are summarized in Table 3. The NO_x in the flue gas mainly existed in the forms of NO and N_2O , and the nitrogen-containing intermediate product mainly existed in the form of HCN. Because the operating temperature of the HTFG was relatively low, NO was mainly derived from the conversion path of fuel NO,³⁰ including the oxidation reactions of the released gas intermediate products (NH_3 and HCN) and char-nitrogen. There were also two conversion paths for N_2O formation.³¹ One was the homogeneous reaction, that is, the NCO radical in released HCN and HNCO from fuel-nitrogen oxidization reacted with NO to form N_2O (Reaction R1), and the other was a heterogeneous reaction (Reactions R2–R3). Meanwhile, the oxygen concentration in the high-temperature flue gas was approximately 10%. The high-temperature flue gas flowed into the main combustion zone and acted as the carrier gas for the pulverized coal in the CC.

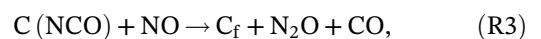
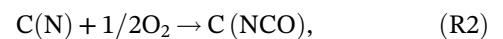
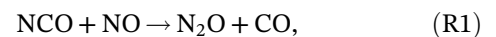


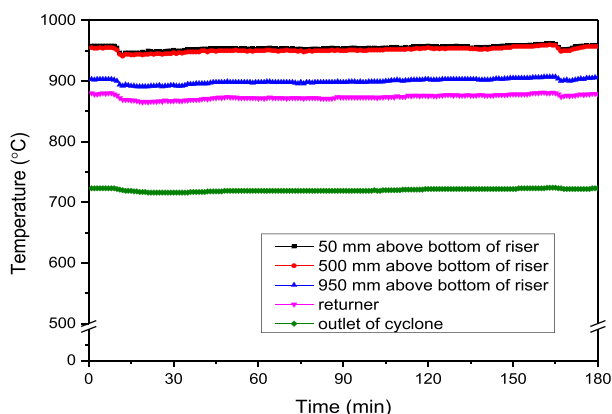
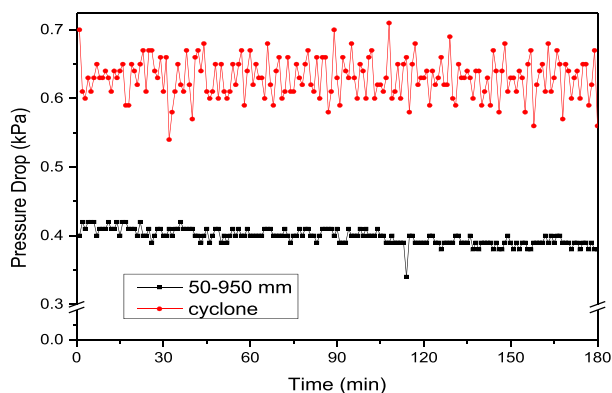
TABLE 1 Proximate and ultimate analyses (wt%, air dried) of fuel

Proximate analysis				Ultimate analysis					Lower heating value
<i>M</i>	<i>A</i>	<i>VM</i>	<i>FC</i>	<i>C</i>	<i>H</i>	<i>N</i>	<i>S</i>	<i>O</i> ^a	<i>Q</i> _{net,ar} (MJ/kg)
8.12	7.68	31.89	52.31	72.31	4.93	0.85	0.30	5.81	24.43

^aBy difference.

TABLE 2 Experimental operating conditions

High-temperature flue gas generator			Combustion chamber				
Coal feeding rate (kg/h)	A_{FA} (Nm ³ /h)	λ_{FA}	Coal feeding rate (kg/h)	A_{PA} (Nm ³ /h)	λ_{PA}	A_{SA} (Nm ³ /h)	λ_{SA}
1.62	25.8	0.21	15.05	65	0.51	64	0.51

**FIGURE 2** Temperature variation in high-temperature flue gas generator**FIGURE 3** Pressure drop variation in high-temperature flue gas generator

where C(N) and C(NCO) refer to nitrogen species on the char surface and an oxidized char nitrogen species (-NCO or -CNO) respectively. C_f refers to a free carbon site.

3.2 | Combustor

The high-temperature flue gas carrying the pulverized coal flowed into the CC and mixed with the primary air

to ignite and burn at the CC entrance. The secondary air was injected at 300, 1,200, 2,400, and 3,000 mm below the top, so that a reducing atmosphere was created in the zone from 0 to 3,000 mm below the CC top, which is conducive to the reduction of NO_x.

3.2.1 | Temperature distribution

Figure 4 shows the temperature variation along the combustion path. The coal particles reached a higher combustion temperature in a short time, and the combustion temperature reached above 1,100°C at a distance of 150 mm below the CC top. Compared with the conventional ignition conditions of pulverized coal, the pulverized coal ignited and burned earlier in this study. This is because the temperature of the high-temperature flue gas stream had exceeded the ignition temperature of the pulverized coal, and the combustion state could immediately be reached when the high-temperature gas–solid mixture was in contact with the primary air. The coal ignition process increased the burnout residence time of pulverized coal in the CC earlier, which was beneficial to the increase in combustion efficiency. At this time, a large amount of volatiles was released in the inlet zone of the CC and filled the zone, and the temperature of the char particles significantly increased. When the first-stage secondary air was injected, a large amount of volatile and char particles began to burn violently, and the temperature in the CC reached its highest value. As the secondary air was injected incrementally, the combustible content in the fuel gradually decreased, and the combustion temperature decreased accordingly. The temperature of the tail flue gas was 619°C.

During stable combustion, the maximum temperature in the CC was approximately 1,181°C, and the temperature difference in the 0–3,000 mm zone along the combustion path was within 230°C. In conventional combustion, the temperature difference in the main combustion zone is usually above 500°C.^{32–34} Therefore, a more uniform temperature distribution was reached in

TABLE 3 High-temperature flue gas composition

NO (ppm)	N ₂ O (ppm)	NH ₃ (ppm)	HCN (ppm)	CO (ppm)	CO ₂ (%)
254.77	11.77	0.21	11.07	279.03	8.37

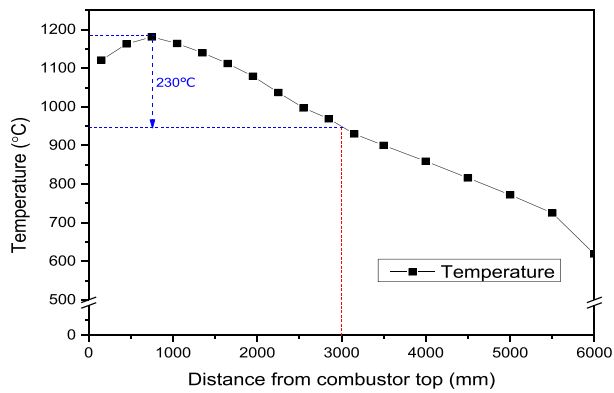


FIGURE 4 Temperature variation along combustion path

the main combustion zone under this combustion mode, and there was no local high-temperature zone in the main combustion zone; the local oxidation zone was avoided. The combustion characteristics are beneficial to the further reduction of NO_x .^{35,36}

3.2.2 | Nitrogen conversion

In this study, the secondary air at different injection locations along the combustion path is defined as SA-1, SA-2, SA-3, and SA-4, respectively. The distribution of the air equivalence ratio in the CC is shown in Figure 5. The CC was divided into five reaction zones, which are defined as Zone I, Zone II, Zone III, Zone IV, and Zone V. The corresponding air equivalent ratios are 0.71, 0.84, 0.97, 1.10, and 1.23, respectively.

With the gradual injection of secondary air, the reaction atmosphere along the combustion path in the main combustion zone gradually changed from a strongly reducing atmosphere to a weak reducing atmosphere. Eventually, the pulverized coal burned out in the oxidizing atmosphere.

The concentration distributions of nitrogen-containing compounds (NO , N_2O , NH_3 , and HCN) and CO along the combustion path are summarized in Figures 6 and 7. The gas concentrations at each position were recorded online every 5 s for 10 min, and the variation in nearly all gas concentrations was within $\pm 10\%$. The results indicate that NO was the main nitrogen-containing compound and the concentrations of other nitrogen-containing compounds (N_2O , NH_3 , and HCN) were extremely low. As a result, this section mainly discusses the variation trend for NO concentration. Combined with the above-mentioned temperature distribution and air-equivalent ratio distribution in the CC, an in-depth analysis of the variation in the NO concentration in several zones is summarized as follows:

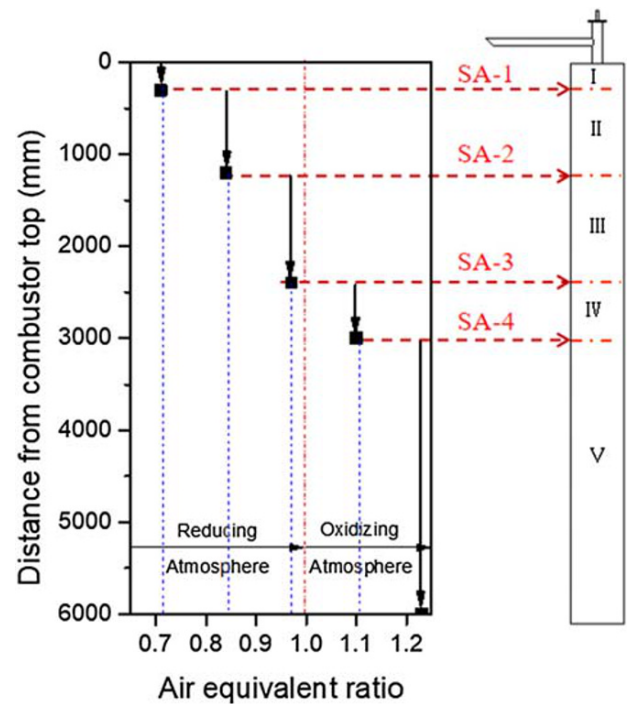


FIGURE 5 Distribution of air equivalence ratio in combustion chamber

Zone I: The pulverized coal carried by the high-temperature flue gas came into contact with the primary air; combustion reactions occurred, and the oxygen was rapidly consumed. Only a small part of the released volatiles was oxidized, and the remaining volatiles surrounded the char particles and filled the zone. At the same time, the CO_2 in the high-temperature flue gas also reacted with the char particles to generate CO in this zone. These reactions led to a sharp increase in the CO concentration from 279 ppm at the entrance to

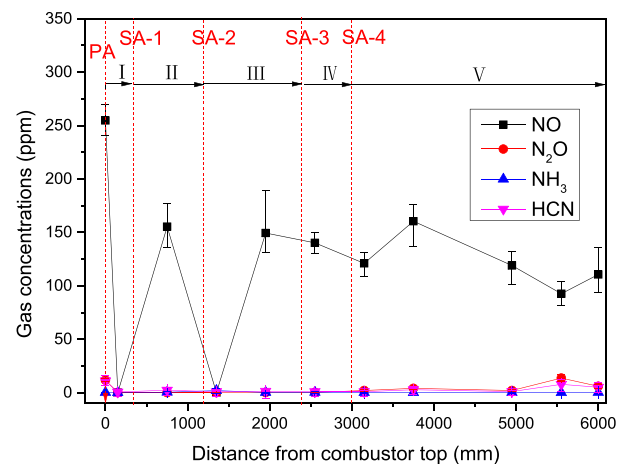


FIGURE 6 Nitrogen-containing gas concentration variation along combustion path

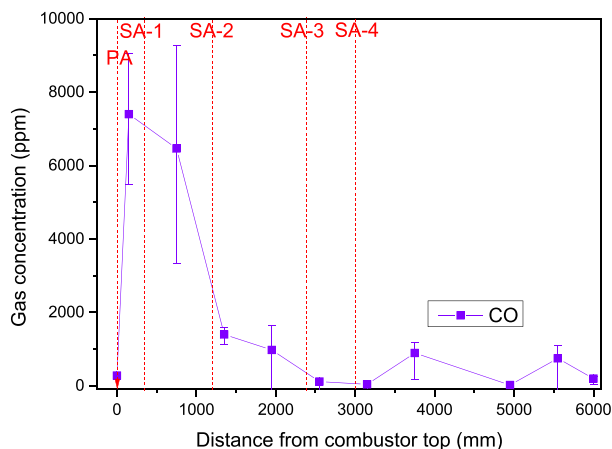
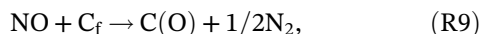
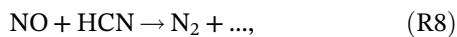
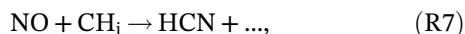
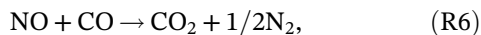
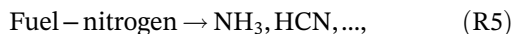


FIGURE 7 CO concentration variation along combustion path

7,399 ppm, indicating that the reaction atmosphere was a strongly reducing atmosphere. In this zone, the chemical reactions of fuel-nitrogen were mainly volatile-nitrogen reactions. Meanwhile, under the catalysis of high-concentration char particles, CO, CH_i, and so forth were used as the main reducing agents to reduce the nitrogen-containing compounds in the volatiles and in the high-temperature flue gas to N₂. The main reactions are summarized as follows³¹:



where C(O) refers to oxygen species on the char surface.

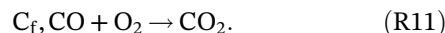
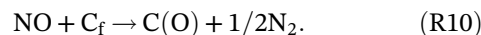
Zone II: With the injection of the first-stage secondary air, the air equivalent ratio in this zone increased from 0.71 to 0.84, and the reducing effect in the reducing atmosphere became weak. At this time, the pulverized coal combustion temperature reached the highest value, indicating that the combustion reaction intensity increased. The release rate of volatiles from coal oxidation and the oxidization rate of volatile-nitrogen to NO both increased. Because the variation in the NO concentration is determined comprehensively by the oxidation and reduction reactions of fuel-nitrogen, the combined effect of the

enhanced oxidation reactions and weaker reduction reactions in this zone increased the NO concentration from 0 to 155 ppm. The concentrations of other nitrogen-containing compounds were basically unchanged.

Zone III: NO concentration in this zone decreased sharply first, then increased to 150 ppm. With the injection of the second-stage secondary air, the air equivalent ratio became 0.97, the CO concentration dropped drastically to 976 ppm, and the reaction atmosphere remained a weakly reducing atmosphere. The high-temperature fuel from Zone II came into contact with the second-stage secondary air to undergo a violent oxidation reaction such that almost all the residual volatiles in the fuel were released. In the early stage of release, a local enhanced reducing atmosphere was created around the char particles,¹⁸ and the catalytic effect that the reactive sites on the char surface had on the reduction reactions of NO also improved. The comprehensive effect contributed all NO in the early stage to be reduced to N₂. With the combustion reactions proceeding, the oxidation reactions of char-nitrogen played a leading role in the variation of the NO concentration, which made the NO concentration increase significantly to 149 ppm.

Zone IV: The reaction atmosphere in this zone transitioned from a weakly reducing atmosphere to an oxidizing atmosphere. The chemical reactions of char-nitrogen played a leading role in the NO concentration. A small amount of oxygen was added to reduce part of the NO,³⁷ which resulted in a slight decrease in NO concentration.

Zone V: This zone was the burnout zone of coal combustion, where the air equivalent ratio was 1.23. During the burnout process of char, the char-nitrogen was completely released, resulting in the local NO concentration increasing to 160 ppm at 3,750 mm below the CC top. Under the heterogeneous reduction of NO on the char surface, the NO concentration gradually decreased, and the NO emissions were 110 ppm. The main reactions are summarized as follows:



During the optimized high-temperature flue gas combustion, the above two reaction paths in Zone I and Zone III, where the NO concentration was reduced to 0 ppm, are the main reasons for the significant reduction in NO_x emissions.

Table 4 shows the gas concentrations of the nitrogen-containing compounds (NO, N₂O, NH₃, and HCN) and carbon-containing compounds (CO and CO₂) in the tail flue gas. Meanwhile, the oxygen concentration in the tail

TABLE 4 Tail flue gas composition

NO (ppm)	N ₂ O (ppm)	NH ₃ (ppm)	HCN (ppm)	CO (ppm)	CO ₂ (%)	O ₂ (%)
110.61	5.92	0.20	5.03	195.38	13.35	3.93

flue gas was approximately 3.93%. The NO_x emissions were mainly NO emissions at a concentration of 110 ppm, while the concentrations of N₂O, NH₃, and HCN were extremely low and could be considered to contribute little to the NO_x emissions. The original emissions of NO_x were only 187.2 mg/m³ (@6% O₂), and the conversion ratio of fuel-nitrogen to NO_x was approximately 16.6%. Compared with the original NO_x emissions (600–800 mg/m³ @6% O₂) in conventional pulverized coal combustion technology,^{33,34} the advantages of this technology route are great.

4 | CONCLUSIONS

In this study, an improved high-temperature flue gas combustion technology was adopted to reduce NO_x emissions. The HTFG generated a flow of flue gas at a high temperature (720°C) with low oxygen content (10%) that flowed into the main combustion zone and acted as the carrier gas for pulverized coal in the CC. In addition, the NO_x in the high-temperature flue gas mainly existed in the forms of NO and N₂O, and the nitrogen-containing intermediate product mainly existed in the form of HCN.

In the CC, the coal particles reached a higher combustion temperature (above 1,100°C) in a short time. Moreover, the maximum temperature in the CC was 1,181°C, and the temperature difference in the main combustion zone was within 230°C. It has been shown that the local oxidation zone was avoided, which is beneficial to the further reduction of NO_x.

Furthermore, air-staging combustion technology was also adopted in combination with this technology. NO was the main nitrogen-containing compound, and the concentrations of other nitrogen-containing compounds (N₂O, NH₃, and HCN) were extremely low. The NO concentrations were reduced to 0 ppm twice when the volatiles released rapidly in the initial stage and the volatiles burned violently in the intermediate stage, which are the main reasons for the significant reduction in NO_x emissions. The original emissions of NO_x were 187.2 mg/m³ (@6% O₂), and the conversion ratio of fuel-nitrogen to NO_x was 16.6%. High-temperature flue gas combustion technology has attained low-NO_x combustion of pulverized coal. The results could provide a theoretical basis for future engineering practices.

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