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Experiment and mechanism investigation on advanced reburning for NO_x reduction: influence of CO and temperature

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Abstract: Pulverized coal reburning, ammonia injection and advanced reburning in a pilot scale drop tube furnace were investigated. Premix of petroleum gas, air and NH_3 were burned in a porous gas burner to generate the needed flue gas. Four kinds of pulverized coal were fed as reburning fuel at constant rate of 1g/min. The coal reburning process parameters including 15%~25% reburn heat input, temperature range from 1100 °C to 1400 °C and also the carbon in fly ash, coal fineness, reburn zone stoichiometric ratio, etc. were investigated. On the condition of 25% reburn heat input, maximum of 47% NO reduction with Yanzhou coal was obtained by pure coal reburning. Optimal temperature for reburning is about 1300 °C and fuel-rich stoichiometric ratio is essential; coal fineness can slightly enhance the reburning ability. The temperature window for ammonia injection is about 700 °C~1100 °C. CO can improve the NH_3 ability at lower temperature. During advanced reburning, 72.9% NO reduction was measured. To achieve more than 70% NO reduction, Selective Non-catalytic NO_x Reduction (SNCR) should need NH_3/NO stoichiometric ratio larger than 5, while advanced reburning only uses common dose of ammonia as in conventional SNCR technology. Mechanism study shows the oxidization of CO can improve the decomposition of H_2O, which will rich the radical pools igniting the whole reactions at lower temperatures.

Key words: NO reduction, Advanced burning, Coal reburning, Selective Non-catalytic NO_x Reduction (SNCR), CO
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INTRODUCTION

Nitrogen oxides (NO_x) are one of the most toxic pollutions formed during the combustion process especially of pulverized coal. International Energy Agency forecasts have indicated that some 38% of the world's electricity will still be generated from coal by 2020 (Priddle, 1998). More efficient and environmental friendly and cleaner coal technologies have therefore a major role to play in sustainable development worldwide.

Nowadays, low NO_x burners for stage-air combustion are widely used in power plant boilers. Combustion modification methods can only achieve moderate NO_x emission level. Without perfect modification, sludge in furnace loss of ignition (LOI) will arise. With further limitation, Selective Catalytic NO_x Reduction (SCR) and Selective Non-Catalytic NO_x Reduction (SNCR) should be employed (Radojevic, 1998). These techniques take advantage of an N-agent to reduce NO to N_2 such as urea or ammonia. SCR requires noble metal catalyst, which is the most efficient but most expensive NO_x emission control technology, nowadays. SNCR does not require catalyst but is difficult to manipulate for the required narrow temperature window (1200~1350 K) in boiler. The most attractive technology proposed recently combined reburning and SNCR process, is called Advanced Reburning (AR) and reportedly could achieve more than 80% NO_x reduction to approach that of SCR at very low cost (Chen and Kramlich, 1989; Tree and Clark, 2000; Hampartsoumian and Folayan, 2003; Nimmo *et al*., 2004); and so, is becoming more attractive for detailed investigation recently.

Reburning is a promising combustion modification technology which had been demonstrated in laboratory reactors and in full scale boilers (Smoot, 1998). In reburning, NO reduction occurs within the furnace by injection of a second fuel in the downstream of the primary combustion zone. The reburning fuel creates a fuel-rich zone where NO can be reduced by hydrocarbon radicals. Air called OFA (over fire air) injected into the downstream of the primary combustion zone burns out the reburning fuel. Gaseous hydrocarbon fuels are more attractive than coal for reburning because NO_x reduction is higher and easy to retrofit in the existing power plant. Additionally, there is no serious burnout problem. However, for most coal-fired utility boilers, pulverized coal is the preferred choice because of its being on-site and low cost.

SNCR process includes Thermal DeNO_x process (ammonia as N-agent), NO_xOUT process (urea as N-agent) and RAPRENO_x process (cyanuric acid as N-agent) (Kasuya and Glarborg, 1995). Because of its simple composition, ammonia is usually used in bench scale test, while urea is mostly used in full scale boilers. Many researches have been done on PSR (perfectly stirred reactor) and plug flow rectors; more than 90% NO_x reduction can be achieved (Kasuya and Glarborg, 1995; Rota and Zanoelo, 2000; Rota and Antos, 2002). Miller and Bowman (1989) and Glarborg and Miller (1994) developed detailed mechanisms for the process which agreed very well with the experimental result. However when applied in full scale boilers, due to the mixing and risk of ammonia slip, SNCR technology can only achieve moderate efficiency for NO_x abatement. It is reported that 40%~50% overall NO reduction could be achieved on pulverized coal boiler (Stallings, 2000), 70% on incineration boiler (Furrer and Deuber, 1998; Zandaryaa and Gavasci, 2001) and 56% on CFB boilers (Ljungdahl and Larfeldt, 2001).

It becomes more complex when the reburning and SNCR process are combined together, especially as the influence of fuel-rich zone on the SNCR process must be investigated in detail. The aim of this work is to investigate the coal reburning process, Thermal DeNO_x process, advanced reburning process and the impacts of CO and temperature on the ammonia-NO_x reaction system.

EXPERIMENTAL SETUP

All measurements were tested in a bench scale drop tube furnace. The reactor shown schematically in Fig.1 is made up of two cylindrical sections 70 mm in diameter and 2.5 m high. Each section has individual electric heating system which can achieve different temperature profiles. A water-cooled probe was inserted into the top sections in order to prevent high temperature oxidization of aqueous ammonia. The aqueous ammonia was directly introduced into the lower section. The premix of petrol gas, air and NH_3 were burned together in a porous burner to simulate the needed primary-zone combustion products in boiler. The simulation flue gas was pumped into the upper furnace. The electricity-heated drop tube furnace simulates the reburning zone. The pulverized coal (considered as reburning fuel) was fed into the furnace about 1 g/min. Reburning heat input was changed with the variation of flue gas. With adjustment of the primary air, atmosphere in the furnace can be changed from fuel-rich to fuel-lean. Aqueous ammonia was injected with a mini pump into the water-cooled probe. As the aqueous ammonia was going down through the higher temperature zone, it was evaporated into gas and injected into the furnace. Gas species of O_2, CO_2, CO, NO, NO_2, SO_2 were monitored continually with online gas analyzer (Testo 350XL).

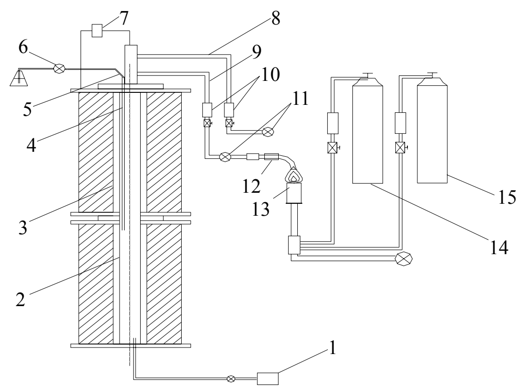

Fig.1 Experimental apparatus schematic

1: Gas analyzer; 2: Corundum tube; 3: Turbo heater; 4: Water-cooling probe; 5: Microfeeder; 6: Mini-pump; 7: Coal feeder; 8: Air; 9: Flue gas; 10: Gas meter; 11: Pump; 12: Suck probe; 13: Porous Burner; 14: Liquefacient ammonia; 15: liquefied petroleum gas

RESULTS AND DISCUSSIONS

Coal reburning process

Four typical coals with different rank around China were selected for reburning in this test. Proximate and elemental analyses of all the reburning coals are given in Table 1. The coal particles were prepared around 80 μm. The air, petrol gas and ammonia flow rates were set by flow-meters and the simulation flue gas NO concentration was kept around 500×10^{-6} at 6% O_2. The total plug-flow gas residence time in the drop-tube was estimated as 1 s for all the experiments. The burn out zone was not incorporated into the system; in this respect it was similar to systems used by other researchers to investigate the fundamental aspects of reburning (Liu *et al*., 1997). Smoot (1998)'s mechanisms of NO reduction with natural gas are listed below:

$$2NO+2CH_4=2HCN+2H_2O+H_2 \quad (1)$$

$$6NO+2CH_4=2CO+4H_2O+3N_2 \quad (2)$$

The mechanism of coal reburning is more complex than that of reburning with natural gas, due to the heterogeneous property of coal, effect of coal rank, composition and fuel-N content. Coal with high volatile and low fuel-N content are usually considered as the favorite reburning fuel. The nitrogen content of the reburning coal partly emitted along with the de-volatilization process is called volatile-N. The volatile-N can also be considered as NO_x precursors including HCN, NH_3, etc. The NO_x precursors have different behavior at different temperature and atmosphere. Liu *et al*.(1997)'s study showed that these NO_x precursors could improve NO_x reduction efficiency in fuel-rich conditions while in the fuel-lean condition contribute to the NO_x emission. When reburning coal particles are injected into high-temperature and high-heating-rate environment, they rapidly release volatile matter within a short time, so that localized fuel-rich pockets are formed around the particles, probably due to the micro scale diffusion around the individual particles. NO can be efficiently reduced by reburning mechanisms in these fuel-rich pockets (Liu *et al*., 1997). The residual fuel-N called Char-N will be oxidized into NO in the following burn out zone. The heterogeneous reactions between NO and coal char are more comprehensive than homogeneous reactions. As the temperature and heating-rate in the combustion chamber is much higher than that in standard proximate test environments, the emission behavior of volatile matter and volatile-N in combustion is usually different for coals with different rank and similar proximate and fuel-N contents.

The selected four coals had similar fuel-N content of 1.09% to 1.30% and different coal rank from bituminous to anthracite. With increasing of reburning heat input, the NO reduction was improved by all the four coals (Fig.2). Yanzhou coal performed best in all tests, probably because it had higher volatile matter content (29.51%) than the others, and so, will emit more C_mH_n radicals during the volatile emission process resulting in efficient NO reduction. But the volatile fraction may not be the only key parameter affecting the reburning performance. Even the Jincheng coal which only has 7.32% volatile fraction achieved 26.3% NO reduction while Yanzhou coal achieved 47% at the 25% reburn heat input condition. That means apart from the contribution of C_mH_n

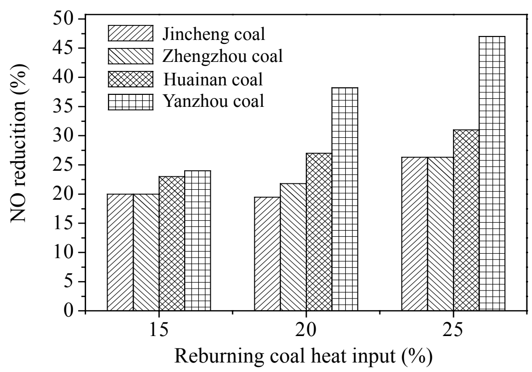

Fig.2 Coal reburning efficiency on NO reduction, *T*=1200 °C

Table 1 Coal properties analysis

Coal	Proximate analysis (wt%)				Ultimate analysis (wt%, daf)				
	Mad	Aad	Vad	FC	Cad	Had	Nad	Sad	Oad
Jincheng	0.52	22.16	7.32	70.00	68.98	3.02	1.09	0.43	3.80
Zhengzhou	0.56	23.65	14.11	61.68	67.00	3.86	1.30	0.72	2.91
Huainan	0.69	29.65	25.45	44.21	60.74	3.82	1.13	0.35	3.62
Yanzhou	1.45	22.94	29.51	46.10	63.86	4.36	1.13	0.58	5.68

radicals, the behavior of volatiles emission, coal rank, nitrogen content, char surface heterogeneous reactions, all contribute to the NO reduction process. Zhengzhou coal with two times the volatile fraction of Jincheng coal had performed similarly in the test; probable because large amount of fuel-N changes into char-N during reburning by anthracite. During the burnout process, char-N contributes to NO emission; Zhengzhou coal's high fuel-N content counteracts the volatile contribution. Huainan coal has the same volatiles like those of Yanzhou coal; but does not have good performance like Yanzhou coal, probable because Yanzhou coal has more moisture and less ash contents. During the first stage of combustion, moisture will consistently be emitted with the volatiles, which form many micro pores in the char, thus increasing the surface for heterogeneous reactions. Little ash could lessen resistance during the volatiles emission process. Therefore Yanzhou coal has the best performance. Fig.3 shows the change of carbon in fly ash when Huainan coal is used as reburning fuel. The reburn zone temperature is about 1200 ºC, residence time is about 1 s. Results showed that with the increase of heat input NO reduction increased from 23% to 31%, while carbon in ash increased form 3.64% to 4.52% which are acceptable values.

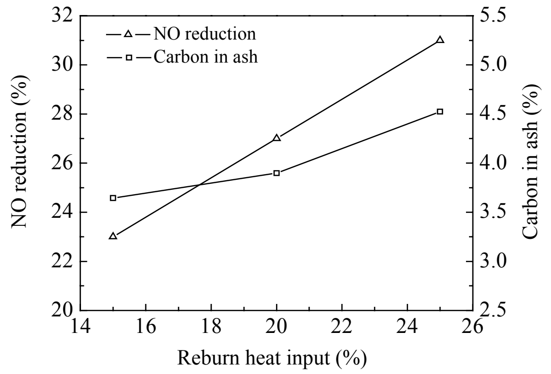

Fig.3 Carbon in ash during the reburning process

Reburn zone temperature is an important parameter of the reburning process. Higher temperature increases the volatile emission in mass and in velocity, which is advantageous for the cracking of char and form more active char surface. The concentration of C_mH_n increases within a short time at higher temperature; which leads to better mixture between C_mH_n and flue gas, therefore leads to increased efficiency. When temperature is high enough, thermal NO_x will become dominant according to the Zeldovich mechanisms. Fig.4 shows that the optimal temperature for the reburn zone is about 1300 °C in this test.

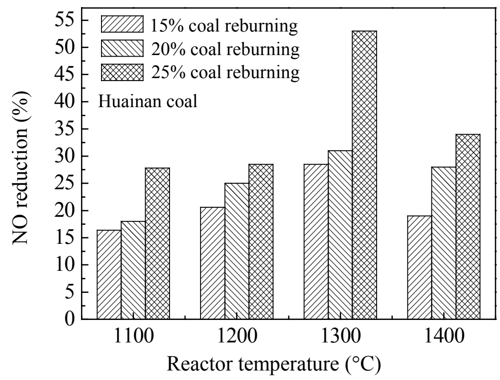

Fig.4 NO reduction with reburn zone temperature

Oxygen in the reburn zone can increase the oxidizing opportunity of C_mH_n radicals and weaken the NO reduction capacity; so that fuel-rich atmosphere is essential as shown in Fig.5. Particle fineness of reburn coal is mostly considered for its burning out property. Fig.6 shows the impact of particle fineness on the NO reduction efficiency. Results showed that at 15% and 20% reburn heat input condition, the impacts of particle fineness on the NO reduction is not so evident. Reburn coal particle size of 40 μm has advantages only at 25% heat input condition. Small particle size of reburn coal is necessary not only for burn out but also for NO reduction. But reburn coal at micro fineness will increase PM10 (particle matter in 10 μm) and PM2.5 emission which are difficult to trap by traditional electrostatic precipitator (ESP). There must be a balanced consideration of the boiler efficiency, NO emission and PM10, PM2.5 emissions level.

Ammonia injection process

Generally, SNCR technology is only capable of achieving a moderate efficiency of NO_x reduction (Stallings, 2000). Although the technology is considered simple to install and operate, it has quite complex chemical reactions and requires strict operation conditions. As an undesirable consequence, SNCR has the problem of ammonia leakage. The ammonia and urea can reduce NO to N_2 at appropriate conditions like with catalyst at 200–400 °C without catalyst at 900–1100 °C (Radojevic, 1998). The reactions are listed below:

$$4NO+4NH_3+O_2=4N_2+6H_2O \quad (3)$$
$$2NO+4NH_3+2O_2=3N_2+6H_2O \quad (4)$$
$$6NO+4NH_3=5N_2+6H_2O \quad (5)$$
$$4NH_3+5O=4NO+6H_2O \quad (6)$$

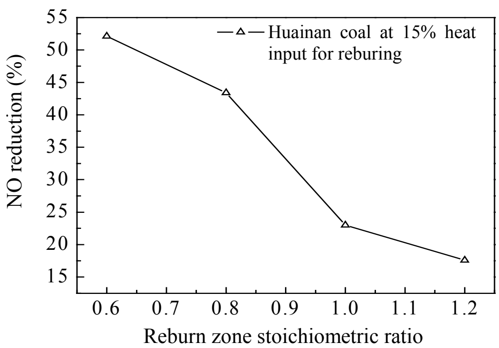

Fig.5 NO reduction with different reburn zone stoichiometric ratio

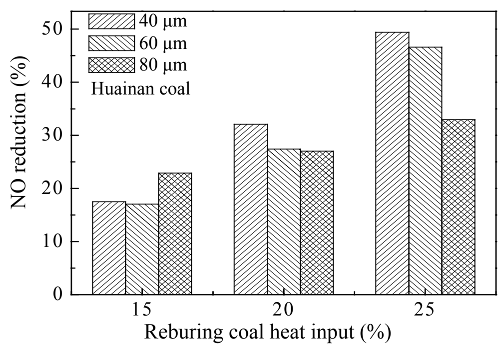

Fig.6 Effect of reburn coal fineness on NO reduction

NH_3 can also be oxidized into NO during the deoxidizing process. The reaction is very sensitive to temperature and environmental condition. Together with the reactor's residence time, there is always a "temperature window" existing. In general, it was reported that 1200~1350 K various from different reactors (Kasuya and Glarborg, 1995; Rota and Zanoelo, 2000; Rota and Antos, 2002).

In this test, saturation ammonia of about 30% was injected from 600 °C to 1300 °C with different NSR (NH_3/NO stoichiometric ratio). Initial gas composition are shown in Table 2. From Eq.(3) and Eq.(4), conclusion can be drawn that the stoichiometric ratio between NH_3 and NO reactions is a value varying from 1 to 2. With increasing of NSR, NO reduction was enhanced under all test conditions, as shown in Fig.7. However, exceeding dose of ammonia will result in risk of ammonia leakage and when using high-sulphur fuels, the formation of ammonium sulphate and bisulphate may foul, clog and corrode equipments. In the full scale boilers, the NSR is always set to no more than 2 (Stallings, 2000). With the influence of gas-ammonia mixing process and temperature fluctuation with boiler load, the NO reduction of only 30%~50% can be achieved. This may be the major obstacle for application of SNCR technologies. Table 2 and Fig.7 show that the temperature window falls into 700 °C to 1100 °C. If the temperature is higher than the temperature window, NH_3 will be oxidized into NO. Suhlmann and Rotzoll (1993) found that the CO can shift the temperature window toward lower temperatures of about 100 K, but that large amount of CO may diminish the NO reduction efficiency. Chen and Lyon (1991) suggested that the optimal CO concentration should be 1500×10^{-6} to 2000×10^{-6}. In this test, more than 1000×10^{-6} CO was added at 700 °C condition, which can be found in Table 2. The same conclusion was obtained but much lower temperature border was observed than others. This might be the major reason why researchers try to combine the reburning with SNCR process.

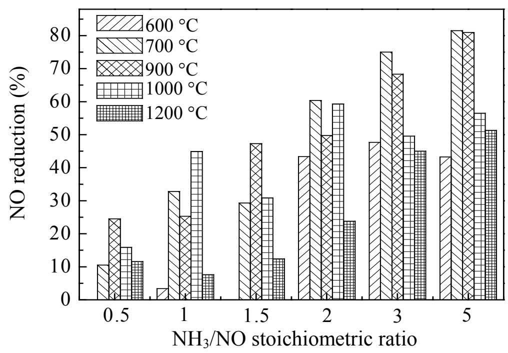

Fig.7 NO reduction with ammonia injection process

Advanced reburning process

In recent years, advanced reburning was introduced and investigated. Chen and Kramlich (1989), and Chen and Lyon (1991) injected N-agent into different zone and temperature. They found that the optimal N-agent injection point with stoichiometric ratio of 0.99 was needed. Zamansky (1997) and Zamansky and Rusli (1999) found that sodium salt promoters could greatly improve the AR performance. Tree and Clark (2000) studied the detailed species profile of the reaction zone. They always used natural gas or methane as the reburning fuel. But in utilizing N-containing pulverized coal, things may be different. According to the different zone of ammonia injection,

Table 2 Flue gas composition of the experiment at *NSR*=3 and residence time *t*=1 s

Temperature (°C)	Before ammonia injection					After ammonia injection					NO reduction (%)
	O_2 (%)	CO (10^{-6})	CO_2 (%)	NO (10^{-6})	NO_2 (10^{-6})	O_2 (%)	CO (10^{-6})	CO_2 (%)	NO (10^{-6})	NO_2 (10^{-6})	
700	8.7	1286	10.8	485	13	8.9	2222	10.6	108	5.6	77.3
850	7.1	210	12.1	466	6.4	8.2	292	11.2	140	5.2	67.3
950	7.2	5	12	549	3.3	7.7	0	11.6	250	1	52.7
1100	2.9	12	15.9	794	4.3	2.5	0	16.3	478	2.1	41.1
1300	4.8	15	14.2	584	8	5.6	0	13.5	472	3.3	22.6

advanced reburning is implemented by two separate processes: AR-rich and AR-lean. In the AR-lean process, ammonia is injected into the burn-out zone with OFA (over fire air) or the downstream of the OFA. The overall efficiency of NO reduction can be better than both reburning and SNCR alone. In the AR-rich process, ammonia is injected into the fuel rich reburn zone with large amount of CO and lesser amount of oxygen. Both of these two characteristics can improve or expand the SNCR process temperature window. Overall NO reduction of 85% was obtained in a down-fired furnace by Hampartsoumian and Folayan (2003), 84% by AR-lean and 93% by AR-rich was achieved in a Boiler Simulator Facility (BSF) by Energy and Environmental Research Corporation (EER, USA) (Zamansky, 1997).

Based on the individual study on coal reburning and SNCR process, coal reburning at 1200 °C and ammonia injection at 950 °C was employed in this test. About 2000×10^{-6} CO and 500×10^{-6} NO were obtained by means of air and petroleum gas premix-flame. Fig.8 shows that 35% NO reduction efficiency was reached with Huainan coal reburning alone. Ammonia injection process can achieve 71.6%

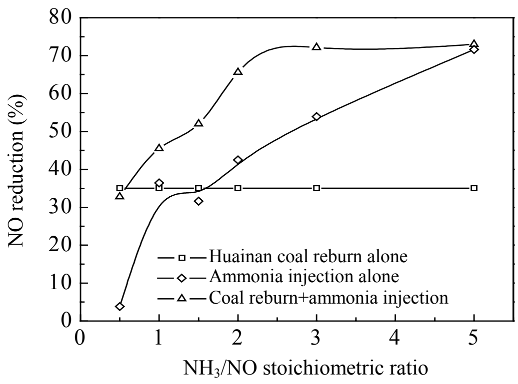

Fig.8 Advanced reburning and SNCR process for NO reduction

NO reduction, while requiring NH_3/NO stoichiometric ratio is larger than 5. Due to the potential risk of ammonia slip, this NSR (larger than 5) should not be used in full scale boilers. In advanced reburning, the NO reduction dramatically increased on the basis of coal reburning before *NSR*<3, 72.1% NO reduction was achieved at *NSR*=3; after that NO reduction stabilized. The results showed that the advanced reburning can feasibly achieve more than 70% NO reduction in utility boilers.

Mechanism studies–influence of CO concentration

Mechanism studies showed that the initial NH_3 is converted to NH_2 by reactions with free radicals such as O, OH, H, etc. The following reactions are be the key steps in the process:

$$NH_2+NO=N_2+H_2O \quad (7)$$
$$NH_2+OH=NH+H_2O \quad (8)$$
$$NH_2+NO=NNH+OH \quad (9)$$

It is a chain-branching reaction; NNH can mostly react with NO, M (medium), OH to form N_2, while NH mainly forms HNO, which will be subsequently oxidized into NO. There are two main routes for NH_3 reaction, one for NO formation and the other for NO reduction. H_2O decomposition and CO oxidization are as described below:

$$CO+OH=CO_2+H \quad (10)$$
$$H+O_2=OH+O \quad (11)$$
$$O+H_2O=OH+OH \quad (12)$$

From which we can find that CO mainly consumes the OH radicals, which will speed up the H_2O decomposition. Large amount of free radicals (O, H) are formed to initiate the whole self-sustained reaction process. The radical pools are dense at higher temperatures, but less dense at lower temperatures. There is competition between CO oxidization and NH_3 decomposition. Table 2 shows that, after the ammonia injection process the CO increased at 700 °C and 850 °C, while at higher temperatures NH_3 and NO all decreased due to the rich radicals. That is why the CO can boost the reactions just at the low temperatures.

CHEMKIN 3.6 was used to study the reaction mechanisms of the influence of CO over NH_3-NO reactions. Twenty-three reactions and 15 species were chosen from Miller and Bowman (1989). They introduced the hypotheses of well stirred reactor and adiabatic closed system. The simulation result can be considered as the ideal status at the experiment conditions. Flue gas composition was chosen from Table 2. Fig.9 shows the free radicals concentration such as NH_2, HNO, H, O, OH, etc. as function of time at 700 °C with or without CO. There are two different stages in Fig.9a, within the first 5 s, large amount of HNO formed, which caused smooth NO reduction. The second stage starts from 5 s, when increasing O, OH increases NH_2 which rapidly reduced NO. Fig.9b with 1286×10^{-6} CO is different from Fig.9a. There is only one stage within 1 s, when large amount of O and OH radicals formed instantaneously, which contributed to the immediate elimination of NO.

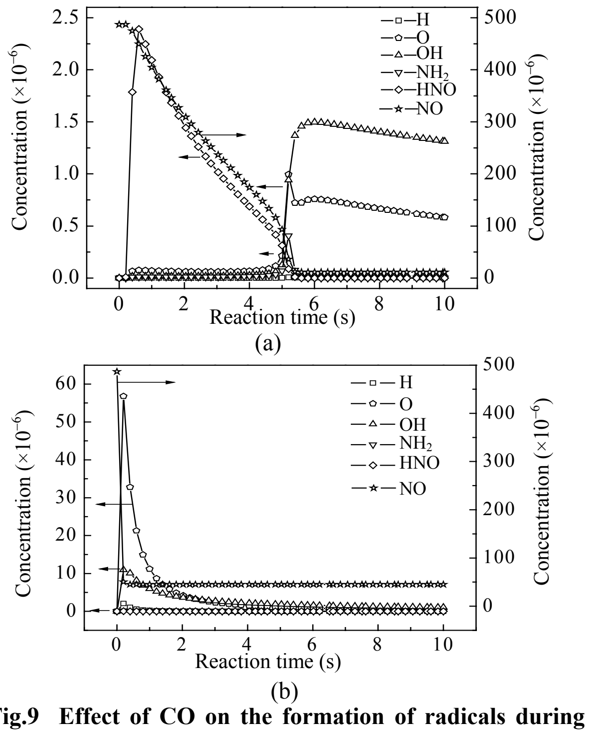

Fig.9 Effect of CO on the formation of radicals during NO-NH_3 reaction process. *T*=700 °C, *NSR*=1. (a) CO=0×10^{-6}; (b) CO=1286×10^{-6}

The amount of CO has remarkable effect on the reaction as shown in Fig.10. With the CO concentration increasing from 0×10^{-6} to 1000×10^{-6}, $[NO]/[NO]_0$ decreases from 0.66 to 0.003, corresponding to 34% and 99.7% NO reductions respectively. From Fig.11a, the CO=1000×10^{-6} line at lower temperature and CO=0×10^{-6} line at higher temperature are the same as those in the experimental result. It proved that homogeneous modeling can give good information about the process. Besides CO, diffusion and mixing may be the important factors during experiments. Compare Fig.11a with Fig.11b which uses more ammonia to weaken the effect of the mixing and dissipation, and minimize the difference between experimental results and simulation.

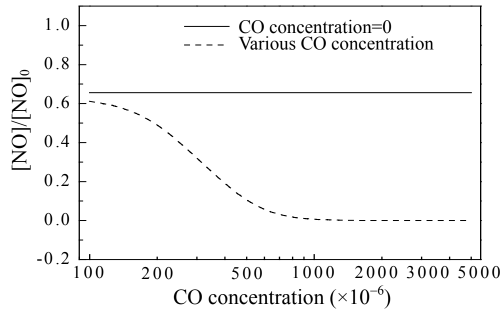

Fig.10 NH_3-NO reaction at different CO concentration. *NSR*=1, *T*=700 °C, *t*=0.5 s

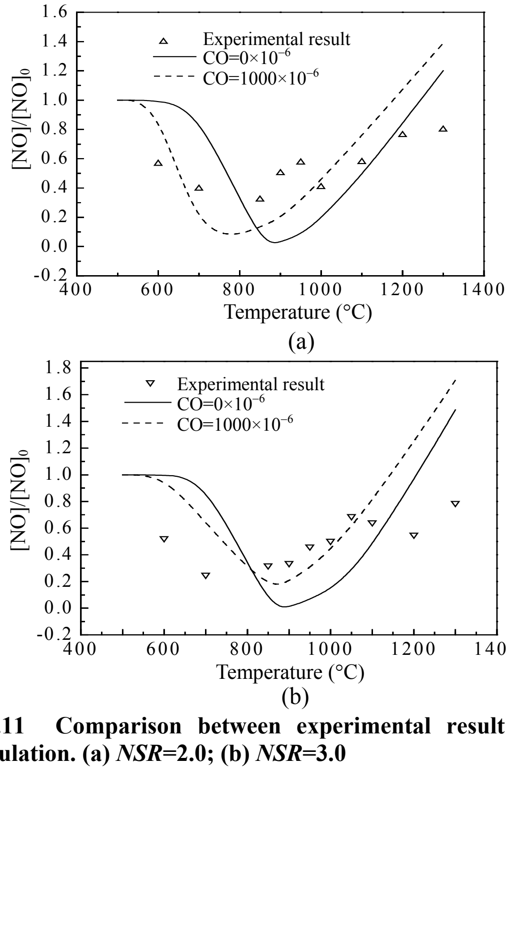

Fig.11 Comparison between experimental result and simulation. (a) *NSR*=2.0; (b) *NSR*=3.0

CONCLUSION

Pulverized coal reburning, ammonia injection and advanced reburning were investigated in a pilot scale drop tube furnace. Premix of petroleum gas, air and NH_3 were burned in a porous gas burner to generate the needed flue gas. The reactor merely acts as the reburn zone, which can avoid the influence of unstable flame and nonuniform temperature profile. Four kind of pulverized coals were fed from the upper section at constant speed of about 1 g/min served as reburning fuel. Ammonia was injected directly or through a water-cooled probe.

NO reduction improved with increasing of reburn heat input. Maximum of 47% NO reduction with Yanzhou coal at 25% reburn heat input was obtained by pure coal reburning. Carbon in fly ash was under the acceptable level. Optimal reburn temperature of about 1300 °C was observed. Coal fineness can slightly improve the coal reburn ability. Fuel-rich reburn zone appropriate stoichiometric ratio can enhance the NO reduction ability. During the pure SNCR process, optimal temperature of about 700 °C to 1100 °C was observed. CO can improve the lower temperature reactions. During advanced reburning, 72.9% NO reduction was observed. Pure Huainan coal reburning can reach about 35% NO reduction efficiency; $NSR<3$ can achieve more than 70% NO reduction in advanced reburning. The pure SNCR process should need $NSR\geq5$ to achieve the same target. Mechanism study was conducted to investigate the influence of CO and temperature. CO can promote the decomposition of H_2O to form more radicals, which can start the whole self-sustaining reactions at lower temperatures. Further investigation on coal char heterogeneous reactions and metal catalyst should be conducted.

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