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# Study on NO heterogeneous reduction with coal in an entrained flow reactor

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ABSTRACT

The effects of coal types with a wide range of volatile matter content including lignite, bituminous coal, and lean coal, as well as the effects of reaction temperature, coal particle size, the primary-zone stoichiometry (SR1) and reburning-zone stoichiometry (SR2), etc. on NO reduction efficiency were carried out systematically in an entrained flow reactor. The heterogeneous NO reduction mechanism was analyzed. The results indicate that the NO reduction efficiencies increase with decreasing SR2 and coal particle size, and with increasing reaction temperature. The char contributions to the total NO reduction efficiency increase with increasing proximate volatile matter, coal particle size, and with decreasing reaction temperature. The char contribution can be reached more than 40% when SR2 is larger than 1.06 or less than 0.92 for XLT lignite. The char contribution at the conditions of SR1 = 1.0 and SR1 = 1.2 is significantly larger than that at SR1 = 1.1 for coals with high-volatile matter at a fixed reburning fraction.

## 1. Introduction

Burning coal produces significant amounts of atmospheric pollutants, such as $SO_2$, $NO_x$ and $CO_2$, etc. Nitrogen oxides ($NO_x$) have been recognized as acid rain precursors that impose a significant threat to the environment. Effective and relatively inexpensive technologies for reducing $NO_x$ emission are based on combustion modification. One of the most promising primary methods of $NO_x$ reduction is reburning, which was originally suggested by Wendt et al. [1]. The reburning technology requires creating three distinct reaction zones along the height of the coal-fired furnace: (1) the primary-zone where about 80–90% of the fuel is burned under fuel-lean conditions and where $NO_x$ are generated, (2) the reburning-zone is the fuel-rich zone into which a reburning fuel (10–20%) is injected to reduce the $NO_x$ produced in the primary-zone to $N_2$, and (3) the burnout zone where additional air is added to complete the combustion of the unreacted fuel. The basic reburning process typically provides 50–60% $NO_x$ control [2].

Natural gas, oil, pulverized coal, and biomass can be effective reburning fuels. However, for most coal-fired utility boilers, pulverized coals would be the preferred choice of reburning fuels because of its availability on site and lower cost [3,4]. Many researchers, such as: Liu et al. [5], Zhong et al. [6], Hardy and Kordylewski [7], Chen and Ma [8] and Chen and Tang [9], studied the effect of the type of reburning fuels on the $NO_x$ reduction and demonstrated that lignites were more effective than natural gas. NO reduction reactions with coals include homogeneous mechanisms involving NO reacting with volatiles and heterogeneous mechanisms involving NO reacting with chars. The homogeneous NO reduction in fuel-rich environment is relatively well understood [10]; however, there has been no systematic investigation of NO reduction by chars of diverse origin and history. Few investigators have studied the role of heterogeneous NO reduction mechanisms during reburning. Chen and Ma [8] undertook reburning experiments in a ceramic flow reactor with simulated flue gas and concluded that heterogeneous mechanisms were more important than homogeneous mechanisms for the lignites, while for bituminous coal, heterogeneous NO-char reactions were essentially negligible as the reburning-zone stoichiometry was >0.75. Liu et al. [5] studied the reburning performance of partly devolatilized char with that of the parent high-volatile bituminous coals and concluded that coals having a high-volatile yield are better reburning fuels than lower-volatile coals. They also proposed a model to determine the relative contributions of volatiles and char to the total NO reduction efficiency.

The primary objectives of this study are to address characteristics of heterogeneous NO reduction during reburning with different types of coals. The effects of coal types with wide volatile matter including lignite, bituminous coal, and lean coal, as well as the effects of reaction temperature in the reburning-zone, coal particle size, the primary-zone air:fuel stoichiometry (SR1), and the overall reburning-zone air:fuel stoichiometry (SR2), etc. on NO reduction efficiency were systematically carried out in an entrained flow reactor. The heterogeneous reduction mechanism was analyzed.

## 2. Experimental

The entrained flow reactor (EFR) was constructed from an alumina tube (50 mm ID × 2.0 m high) as shown in Fig. 1. It consists

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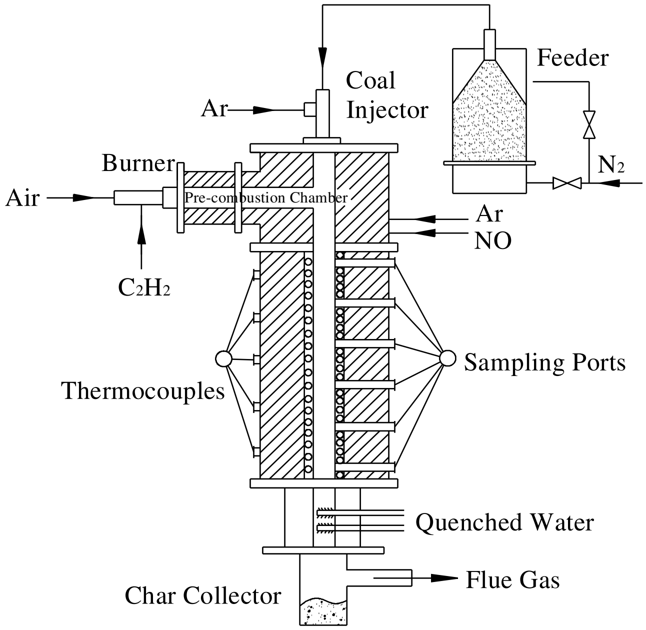

**Fig. 1.** Schematic diagram of entrained flow reactor.

of a primary-zone, a reburning-zone with an electrical heating system, a fluidized bed powder delivery system, a gas supplying system, a temperature measuring and indicating system, a char quenching and collecting system, and a flue-gas sampling and analysis system. A fluidized bed particle delivery system was used to supply the reburning coal pneumatically (using nitrogen as carrier gas) through a water-cooler injector. An acetylene-fired nozzle-mix burner was used to simulate the primary-zone of reburning process. NO was added to the downstream of pre-combustor to simulate the primary-zone combustion products of the reburning processing, in which NO concentration was kept constant at 1000 ppm. Three different primary-zone air:fuel stoichiometric ratios which are SR1 = 1.0, 1.1, and 1.2, were principally used in the experiments. The reburning-zone air:fuel stoichiometric ratios (SR2) were decided by the coal feed rate and primary-zone combustion conditions. The gas temperature within the EFR was controlled at 1000 ± 20 °C and 1200 ± 20 °C, respectively. The total plug-flow gas residence time in the EFR was estimated at 800 ms based on the gas flow rate and the reactor volume for all experiments. Gas and solid samples could be taken from any one of the six sampling ports along the EFR using a short water-cooler 5 mm (ID) stainless probe. All concentrations of $NO_x$, CO, $CO_2$, and $O_2$ were measured by an on-line analyzer on a dry basis. Solid particles were quenched and trapped in water, filtered and dried before analysis. The coal mass loss and the release fraction of carbon, hydrogen, nitrogen in coals were quantified by the ash-tracer method based on analyses of solid samples [11].

Three Chinese coals varying from lignite to lean coal and their chars produced under different conditions were selected as the reburning fuels. The proximate and ultimate analyses of the test coals are shown in Table 1. The reburning experiments were carried out with particles in the size range of $d_p$ = 0–125 μm, which were separated into two fractions: coarse coal particles (75–125 μm) and fines (0–75 μm). The particle sizes of partly devolatilized chars were crushed and screened to less than 75 μm. The reported NO reduction efficiency was calculated as:

$$\eta_{NO} = (1 - [NO]/[NO_0]) \cdot 100\%, \tag{1}$$

where $\eta_{NO}$ is the NO reduction or reburning efficiency, $[NO_0]$ is the primary-zone NO concentration, and [NO] is the NO concentration at the bottom of reactor during reburning. $NO_2$ concentrations were found to be <5% of total $NO_x$ for all conditions investigated.

**Table 1**
Analyses of the coals used in the experiments

| Coal sample | Proximate analysis (wt%) | | | Ultimate analysis (wt%) | | | | |
|---|---|---|---|---|---|---|---|---|
| | $M_{ad}$ | $A_d$ | $V_{daf}$ | $C_d$ | $H_d$ | $N_d$ | $O_d$ | $S_d$ |
| Xiao Long Tan lignite | 22.82 | 8.25 | 43.38 | 65.15 | 2.88 | 1.56 | 21.21 | 0.96 |
| Huai Nan bituminous coal | 1.62 | 16.73 | 32.92 | 70.67 | 4.26 | 1.17 | 6.22 | 0.95 |
| Liu Zhi lean coal | 1.48 | 14.15 | 14.99 | 76.48 | 3.34 | 1.20 | 1.87 | 2.95 |

Three sets of tests, which included char preparation, coal reburning, and char reburning, were carried out under similar experimental conditions. To analyze the contributions made by chars to the total NO reduction, $\eta_{char}$, it was assumed that the coals consisted of both chars and volatiles. Volatiles contributed to the homogeneous NO reduction during reburning, while reburning with chars revealed maximum heterogeneous NO reduction under no competition from volatiles [8]. Therefore, when the NO reductions from char reburning were compared with those from coals reburning of equivalent char feeding rate (or SR2); the contribution of the heterogeneous mechanisms was clearly shown.

## 3. Results and discussion

### 3.1. Effects of coal types and reburning-zone stoichiometry on NO reduction efficiency

Fig. 2 illustrates the typical effect of the reburning-zone air:fuel stoichiometry on NO reduction efficiency of three different coals and their chars, in which the tested chars were made under the conditions of coal particle size of 0–75 μm, temperature at 1200 °C, SR1 = 1.2, and gas residence time at 800 ms. It can be seen that the NO reduction efficiency is almost inversely proportional to the reburning-zone stoichiometry (SR2) for a constant primary-zone stoichiometry (SR1). For all tested coals and their chars, the NO reduction efficiency of coals having a high-volatile matter is higher than that of lower-volatile coals at the same reburning-zone air:fuel stoichiometry (SR2), which indicates that coals having a

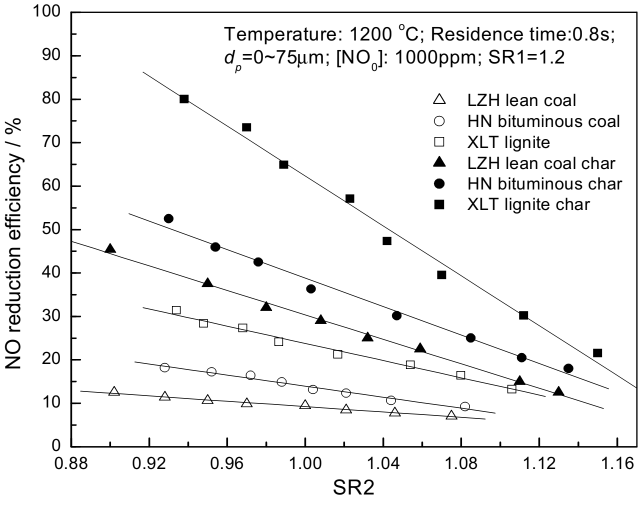

**Fig. 2.** Effect of reburning-zone air: fuel stoichiometry on NO reduction efficiency.