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Research article

# Experimental study on $NO_x$ reduction from staging combustion of high volatile pulverized coals. Part 2. Fuel staging

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#### ABSTRACT

This paper focused on NO<sub>x</sub> reduction from high volatile pulverized coals by fuel-staged combustion, especially from low rank coal in China with strong slagging tendency, high ash, high moisture, and low calorific value. The results are very important to further enrich the database of fuel-staged combustion. The influences of the main process parameters, including reburn fuel fraction, stoichiometric ratio, residence time in reduction zone, and fuel properties, on NO<sub>x</sub> emissions were studied experimentally using an Entrained Flow Reactor with Multiple Reaction Segment (EFRM). The present experiments verified that the comprehensive NO<sub>x</sub> reduction index ( $S_z$ ) proposed by the authors for air-staged combustion is still applicable to correlate the maximum NO<sub>x</sub> reduction rate and coal characteristics in fuel-staged combustion, which would help to predict NOx emissions from fuel-staged combustion more accurately. The experiments also verified that the NO<sub>x</sub> reduction rate increases with the increase of reburn fuel fraction, with the increase of residence time in reburn zone, and with the decrease of stoichiometric ratio in reburn zone. The results also showed that these parameters have a critical range value, which is 15%–20% for reburn fuel fraction, less than 0.8 for reburn zone stoichiometric ratio, and less than 0.8 s for residence time in reburn zone, respectively. NO<sub>x</sub> emissions drop with the decrease of the stoichiometric ratio in main combustion zone until it reaches 0.8. For high volatile coals, the effect of fuel-staged combustion on the reduction of NO<sub>x</sub> emissions increases as the volatile content or fuel-N in the coal increases. The  $NO_x$  reduction rate increases while the comprehensive  $NO_x$  reduction index goes up. Moreover, fuel-staged combustion did not significantly reduce the burnout rate of the high volatile coal.

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#### 1. Introduction

NO<sub>x</sub> emissions from coal combustion are significant contributors to atmospheric pollution, which are responsible for global warming, ozone depletion, acid rain, photochemical smog, and even particulate matters (PM<sub>2.5</sub>) transformation [1–3]. The air-staged and/or fuel-staged combustion technologies, which are the-state-of-the-art technologies for reducing NO<sub>x</sub> formed from coal fired power plants, are widely applied to NO<sub>x</sub> reduction. The NO<sub>x</sub> reduction rate achievable with air-staged combustion is around 70% when using Chinese high volatile pulverized coals (including low rank coal) [4]. However, as NO<sub>x</sub> emissions are further more strictly controlled in China [5], intensive research on NO<sub>x</sub> reduction techniques becomes more important.

Fuel-staged combustion or reburning, different from the air-staged combustion technology, is presently considered to be one of the most promising methods of reducing  $NO_x$  emissions. Proposed by Wendt et al. [6] as far back as 1970s, fuel-staged combustion has been proved

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http://dx.doi.org/10.1016/j.fuproc.2015.06.019 0378-3820/© 2015 Elsevier B.V. All rights reserved. to be a relatively effective and economical combustion technology. By fuel-staged combustion,  $NO_x$  removal greater than 50% may be obtained. Because it has been rapidly accepted as a practical technology for reducing  $NO_x$  emissions, a large amount of researchers [7–22] have been attracted, whose studies have shown that fuel-staged combustion can effectively reduce  $NO_x$  emissions.

Although hydrocarbon fuels can all be used as reburn fuel [10-12], it is more reasonable and attractive for coal-fired power plants to use pulverized coal, especially of high volatile, as reburn fuel. Studies [13-16] have demonstrated that pulverized coal as reburn fuel significantly reduces NO<sub>x</sub> emissions without requiring more expensive fuels. Also, a number of prior researchers [17,18] have shown that coal volatile content plays an important role in fuel-staged combustion in that the amount of hydrocarbon radicals generated increases with the increases of the coal volatile content, resulting in a higher reduction rate of NO<sub>x</sub>. Meanwhile, long reburn zone residence time, small coal particle sizes, and good mixing conditions are vital to fuel-staged combustion. In addition, some studies [14,19,20] indicated that better NO<sub>x</sub> reduction by fuel-staged combustion was achieved with subbituminous coal and lignite than with bituminous coal. The NO<sub>x</sub> reduction 2

### **ARTICLE IN PRESS**

J. Yang et al. / Fuel Processing Technology xxx (2015) xxx-xxx

rate can reach as high as 50% to 70%, when reburn fuel fraction is over 20%. With two lignites and a bituminous coal, the effect of different pulverized coals and chars on NO<sub>x</sub> heterogeneous reduction in reburn zone was studied by Zhong et al. [21,22]. The results indicated that better NO<sub>x</sub> reduction was achieved with lignite and lignite chars than with bituminous coal and bituminous coal chars, and the experimental results also showed that heterogeneous mechanism, coal volatile content, surface area of char particles and char metal oxide content significantly affect NO<sub>x</sub> reduction. Actually, the influences of various physical and chemical characteristics of coal, including volatile content, char specific surface area and the like, on NO<sub>x</sub> reduction are correlated with each other, such correlations have been investigated by a few studies. Generally, the main idea of previous studies on NOx reduction in fuel-staged combustion process is not to control NO<sub>x</sub> emission in the primary (main) combustion zone, which is in oxidizing or weakly reducing atmosphere, then the NO<sub>x</sub> formed upstream is reduced with reburn fuel in the reburn zone. The oxidizing or weakly reducing atmosphere in main combustion zone may cause a large amount of NO<sub>x</sub> to generate at the outlet of main combustion zone, increasing NO<sub>x</sub> reduction burden at the inlet of reburn zone and making it difficult to further reduce NO<sub>x</sub>. By contrast, we proposed a modified reburn system by setting a reducing atmosphere in the main combustion zone to effectively inhibit the formation of NO<sub>x</sub> from the initial stage, which is very significant to ensure a lower final NO<sub>x</sub>. Furthermore, those studies mainly focus on bituminous coal and a small amount of high quality lignite, which is of better coal characteristics, i.e., of low moisture, low ash and high calorific value. Few studies focused on NO<sub>x</sub> reduction by fuel-staged combustion from low rank coal, especially from the lignite and sub-bituminous coals in China with high moisture, high ash, low calorific value and strong slagging tendency. According to WEC statistics [23], China's annual output of lignite is second only to Germany in the world in 2011. Furthermore, over 55% of China's proven coal reserve is low rank coal [24]. Considering the growing crisis of air pollution and the distribution of China's reserves of lignite with high volatile and high moisture as well as lessons learned from developed countries, we believe that developing cost effective technologies to reduce NO<sub>x</sub> emissions from coals for China is one fundamental method to reduce NO<sub>x</sub> emissions. Therefore, it is well worth studying the NO<sub>x</sub> emissions from burning China's low rank coal in fuel-staged combustion.

This paper focuses on the control of  $NO_x$  emissions in modified fuel-staged combustion with four lignites, one sub-bituminous coal and one hv-bituminous coal, which have very high volatiles, and low net heat value. In particular, the impacts of combustion parameters and coal types are examined. Moreover, the theory of grey systems [25] is used to forecast the results of  $NO_x$  reduction in fuel-staged combustion in terms of the physical and chemical properties of coal and char. Experiments were carried out at the CERI-HIT's Entrained Flow Reactor with Multiple Reaction Segment (EFRM). The results have great reference values for the in-depth understanding of the reductions of pollutants emitted from staging combustion of low rank coal in China and the optimization of Chinese existing coal-fired units with high volatile coal. In addition, the results are also important to further enrich the database of fuel-staged combustion.

#### 2. Experimental apparatus

#### 2.1. Experimental facility

The schematic diagram of the electrically heated, Entrained Flow Reactor with Multiple Reaction Segment (EFRM) is presented in Fig. 1, which has a 3.1 m long reaction tube made of corundum-mullite ceramic with an inner diameter of 175 mm. The wall temperatures of the reactor may be set in seven regulated heating zones, with the maximum temperature up to 1600 °C. The pulverized coal scraper feeder at the top end of the furnace is the feeder of main combustion zone, with a pulverized coal mass flow between 0 and 5 kg/h. Two smaller scraper feeders (0-1 kg/h) are added at the appropriate

locations in the furnace in order to supply a constant coal feeding rate of reburn fuel. The details of the facility are summarized in Table 1. The facility can be used to effectively simulate the vertical one-dimensional combustion condition in an actual boiler and to study the effects of fuel-staged combustion on the reduction of  $NO_x$  emissions. A more detailed description of the facility was given in Ref. [4].

#### 2.2. Experimental conditions

In order to compare with air-staged combustion [4], the following experimental studies on NO<sub>x</sub> reduction of the high volatile pulverized coal with fuel-staged combustion are executed. For each experiment, the furnace wall temperature was kept at a constant value of 1250 °C in the reduction zone (main combustion zone and reburn zone) and 1200 °C in the burnout zone. With the total thermal input kept at a constant value of 9 kW, the total coal feed rate in each experiment increases while the net calorific value (as-received basis) of fuel decreases. In fuel-staged combustion, the impact of reburn fuel fraction  $(R_{\rm ff})$ , the stoichiometric ratio in main combustion zone  $(\lambda_1)$  and in reburn zone ( $\lambda_2$ ), the residence time in reburn zone, and the coal characteristics on the nitrogen oxide reduction are studied. Six Chinese coals with different ranks were used for the experiment, including four lignites, one sub-bituminous coal and one hv-bituminous coal. They are Yuanbaoshan lignite, Baiyinhua lignite, Yimin lignite, Xiaolongtan lignite, Zhundong sub-bituminous coal, and Shenhua bituminous coal, which are denoted as YBS, BYH, YM, XLT, ZD, and SH respectively hereinafter. All coals except XLT came from the corresponding coal mining area in the north part of China, XLT lignite came from the Xiaolongtan mining area, Yunnan province in the southwest of China. The analyses of their characteristics are shown in Tables 2–4. The experimental conditions are summarized in Table 5.

The analysis in the part of the Results and discussion section was based on Eqs. (1)–(4) [4,26–30]. Eq. (1) shows the relationship between NO<sub>x</sub> reduction rate and NO<sub>x</sub> emissions in the furnace exit. Eq. (2) is the conversion rate of fuel-N. The elemental release rate ( $\eta_k$ ) and burnout ratio ( $\eta_b$ ) of coal can be expressed by Eqs. (3) and (4), respectively.

$$\eta = 100 \big( 1 - [\text{NO}_{\text{out}}] / \text{NO}_{\text{un-staged}} \big) \tag{1}$$

$$\xi = \left(10^{-2} [\text{NO}_{\rm x}] V_y / 22.4\right) / (N_{ad} / 14) \tag{2}$$

$$\eta_k = 100(1 - (k_1 A_0) / (k_0 A_1)) \tag{3}$$

$$\eta_b = 100((1 - A_0/A_1)/(1 - A_0)) \tag{4}$$

where  $\eta$  is the NO<sub>x</sub> reduction rate (%).  $\xi$  is the conversion rate of fuel-N to NO<sub>x</sub> (%). [NO<sub>out</sub>] and [NO<sub>un - staged</sub>] are NO<sub>x</sub> emissions at the furnace exit during fuel-staged combustion and un-staged combustion (mg/m<sup>3</sup> converted to 6% O<sub>2</sub>), respectively. [NO<sub>x</sub>] is the NO<sub>x</sub> emissions at the furnace exit (ppm).  $V_y$  is the flow rate of dry gas per kg feeding rate of coal (Nm<sup>3</sup>/kg).  $\eta_k$  is the release rate of element k (k = C, H and N) (%), and k is the weight fraction of element k in the sample ( $k_1$ ) or coal ( $k_0$ ).  $\eta_b$  is the coal burnout rate based on ash (%),  $A_0$  is the weight fraction of ash in the sample.

#### 3. Results and discussion

#### 3.1. The effect of reburn fuel fraction

The ratio of the thermal input from the reburn fuel to that from the total fuel (amount of total fuel = amount of main fuel + amount of reburn fuel) is defined as reburn fuel fraction ( $R_{ff}$ ) of fuel-staged combustion [8,12,31]. As shown in Fig. 2, NO<sub>x</sub> reduction rate is effectively

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