Combustion and NO\textsubscript{x} emissions in deep-air-staging combustion of char in a circulating fluidized bed

Zhiqiang Gong *, Zhentong Wang, Lei Wang and Aixun Du

College of Chemical Engineering, China University of Petroleum (East China), Qingdao, China

*Corresponding author e-mail: gzhiq@163.com

Abstract. Combustion and NO\textsubscript{x} emissions in deep-air-staging (with higher level secondary air (SA) injection) combustion of char have been investigated in a CFB test rig. A good fluidized condition and uniform temperature distribution can be achieved with injection of higher level SA. NO\textsubscript{x} emission decreases with injection of higher level SA and the reduction effect is more obvious at higher temperature. NO\textsubscript{x} emission decreases with combustion temperature increasing for char combustion.

1. Introduction

Nitrogen oxides (NO\textsubscript{x}) emitted from coal-fired power plant is a serious environmental issue for years worldwide. Currently, with the increasing deterioration of the environment, NO\textsubscript{x} emission standards become much more stringent in many countries, for example, the limit allowed for power plants is 100 mg/m\textsuperscript{3} at 6\% O\textsubscript{2} from 2014 in China. NO\textsubscript{x} reduction techniques from coal combustion process have attracted great attention.

Circulating fluidized bed (CFB) combustion technology has received great interest and made great progress over the past two decades [1-3]. Compared with traditional pulverized coal combustion, CFB has a great advantage in low NO\textsubscript{x} emission. However, this advantage is weakened under stringent emission limit. NO\textsubscript{x} emission from CFB combustion depends on many operating parameters, among which air stoichiometric ratio ($\lambda$) and secondary air (SA) ratio in air staging combustion are of great importance. Besides, some researchers have studied the effect of SA position [4-7] on NO\textsubscript{x} emission. Wang [4] studied the impact of SA ratio and position on the NO and N\textsubscript{2}O emission and first revealed the axial distribution of NO and N\textsubscript{2}O in the furnace. Ersoy [5] have studied the influence of SA injection on the hydrodynamics of CFB test rig and found that a dense turbulent zone and a relatively dilute bed above the injection port were divided by the SA. Koksal [6] have investigated the effects of air staging on wall-to bed heat transfer and pointed that heat transfer depends strongly on the cross-sectional average suspension density for both SA and non-SA operation.

It has been proved that the air staging technique can significantly reduce NO\textsubscript{x} emission but variations of SA position have little effect on further NO\textsubscript{x} reducing. However, most researchers changed the SA position in the dense area of the riser, the study of effect of SA height along the whole furnace (deep-air-staging) on the hydrodynamics, combustion and NO\textsubscript{x} emission in coal combustion is not sufficient. This paper focused on the effect of SA arrangements (heights of SA port from the distributor plate) along the whole furnace on the hydrodynamics, combustion and NO\textsubscript{x} emission in
char combustion in a CFB test rig. The goal of this paper is prepared for seeking super-low NOx emission solutions.

2. Experimental section

2.1. Fuel
Coal char was made from slow pyrolysis of a sub-bituminous coal at relatively low temperature from 600 °C to 700 °C. After pyrolysis, char was sieved into different particle sizes. Proximate and ultimate analyses of char are listed in Table 1. Compared with the parent coal, the volatile content of char decreases significantly and the fixed carbon and ash contents increase obviously. The diameters of char are 0-4 mm with 50% cut mean diameter (d50) of 2.90 mm.

| Items                     | Char          |
|---------------------------|---------------|
| Proximate analysis (wt%)  |               |
| Moisturea                 | 14.60         |
| Volatile matterb          | 9.89          |
| Fixed carbona             | 66.72         |
| Asha                      | 11.36         |
| Ultimate analysisa (wt%)  |               |
| Carbon                    | 68.31         |
| Hydrogen                  | 0.85          |
| Oxygen                    | 4.01          |
| Nitrogen                  | 0.58          |
| Sulfur                    | 0.30          |
| Low heating valuea (MJ/kg)| 23.32         |

a As received.
b Dry and ash free basis.

2.2. CFB test rig
Experimental tests of char were carried out in a CFB test rig. A schematic diagram of the test rig is demonstrated in Fig. 1, which is composed of a CFB and an auxiliary system. The CFB furnace adopts heat insulation structure with a diameter of 430 mm and a height of 9 m. The recirculation loop is combined of a cyclone, a pneumatic distribution valve, an external heat exchanger (EHE), a loop seal, and two screw feeders. The preheated primary air (PA) is fed into the furnace at the bottom through the air distribution plate, and preheated SA is fed through the ports arranged at four different heights of 1.2 m, 2 m, 4 m and 6 m above the air distribution plate. There are two symmetrical SA ports at first and second level, respectively. Only one SA port is arranged at third and fourth level, respectively. Each SA port is designed to make the SA enter down into the furnace at an angle of 45°, which can make SA a long effective injection distance, pass through the center of horizontal plane [5], resulting in a good gas-solid mixing performance.

There are seven thermocouples and five pressure transmitters along the furnace. Three sampling ports are set: two are in the back pass duct for sampling fly ash and flue gas, respectively; and one is at the bottom slag port for sampling bottom ash. The gas is sampled, dried and filtered before entering individual online analyzers. Gasmet FTIR DX-4000 analyzer and KM9106 portable flue gas analyzer
are used for analyzing flue gas. Oxygen content in flue gas is monitored online by a CY-IS-G zirconia oxygen analyzer in the back pass duct.

Figure 1. Schematic diagram of the CFB test rig.

Table 2. Operation conditions of the experiments.

| Cases | Combustion temperature | $\lambda$ | $\beta$ | SA injection arrangement |
|-------|-------------------------|-----------|--------|-------------------------|
|       |                         |           |        | 1st level | 2nd level | 3rd level | 4th level |
| 1     | 872°C                   | 1.15      | 52%    | on        | off       | off       | off       |
| 2     | 874°C                   | 1.18      | 52%    | on        | off       | on        | off       |
| 3     | 873°C                   | 1.20      | 50%    | on        | off       | on        | on        |
| 2.1   | 925°C                   | 1.22      | 51%    | on        | off       | off       | off       |
| 2.2   | 926°C                   | 1.21      | 50%    | on        | off       | on        | off       |
| 2.3   | 923°C                   | 1.22      | 51%    | on        | off       | off       | on        |
| 3.1   | 936°C                   | 1.22      | 56%    | on        | off       | off       | off       |
| 3.2   | 948°C                   | 1.30      | 49%    | on        | off       | on        | off       |
| 3.3   | 943°C                   | 1.18      | 53%    | on        | off       | on        | on        |

2.3. Experimental conditions

The experimental conditions are listed in Table 2. Each test lasted more than one hour stably. About 70 kg silicon sand with the particle size of 0-1 mm was added into the furnace before the experiments, establishing ash circulation and making the fuel to be fluidized well in the furnace. In the following discussion on the results, NO$_x$ concentration has been normalized to flue gas with dry and oxygen concentration of 6%. Combustion temperature (represented by the highest temperature in the furnace for each case) was different for Cases 1, 2 and 3, 870°C, 920°C, and 940°C, respectively. Air stoichiometric ratio $\lambda$ was maintained at 1.20, SA ratio $\beta$ was maintained at 50% and the first level SA was injected and second SA was not injected for all experimental cases. There are three SA injection arrangements for Cases 1, 2 and 3. As mentioned above, the status of the first and second SA would not be changed, therefore, the variation for each case is the status of the third and fourth SA. Detailed information can be seen in Table 2. In order to develop and maintain a good fluidized state in the
furnace, the PA and the first level SA were injected in all the experiments and the amount of PA and the first level SA should ensure the superficial gas velocity was larger than 3 m/s in the primary region, under this circumstance, the amount of third and fourth level SA was 20%-30% of the total amount of air.

3. Results and discussion
Pressure distribution and distribution of air stoichiometric and superficial gas velocity along the furnace for various SA injection arrangements are shown in Fig. 2 and 3, respectively.

Fig. 2 shows that as the higher level SA injects, there is an increased pressure in the dense region while the pressure in the upper region is almost the same. The higher pressure represents a larger solids holdup in the dense area as the injection of higher SA. With the third or fourth level SA injecting into the furnace, the amounts of PA and first level SA in the dense region decrease as the amount of the total air is constant for each case in Cases 1, 2 and 3, respectively. Therefore, solids carrying capacity in the dense region decreases, resulting in an increasing solids holdup and pressure in the dense region. The difference in pressure is decreasing at higher position as third or fourth level SA injecting. From Fig. 3, it can be seen that a good fluidized condition has been established in all cases, and the influence of higher level SA injection on solids holdup is not significant.

Fig. 3 shows the distribution of air stoichiometric $\lambda$ and superficial gas velocity along the furnace for various SA injection arrangements. The air stoichiometric $\lambda$ is about 1.20 in the exit of the furnace in all experiments. When the third and fourth level SA inject, $\lambda$ is between 0.75 and 0.92 below 2 m and increases to a value between 0.92 and 1.03 below 4 m. While for cases of no injection of the third and fourth level SA, $\lambda$ is larger or near 1.00 above 1.2 m (the height of the first level SA). Obviously, the reduction region is significantly lengthened in the furnace when higher SA injects, which can avoid much NOx produced from oxidation of nitrogen-containing precursors in the primary region and strengthen the NOx reduction reactions in the upper region.

Distribution of superficial gas velocity is uniform for cases of no injection of the third and fourth level SA. The lowest gas velocity appears at the lowest region in the furnace while it becomes larger when the first level SA injects and maintains the value to the upper region. For cases of injection of the third and fourth level SA, gas velocity firstly decreases as the sudden expansion of the cross area at the height of 1.2 m, as can be seen in Fig. 1 and then increases above 4 m or 6 m as the injection of the third or fourth level SA. The lowest gas velocity is 2.80 m/s in Case 3.3, which can develop the basic fluidized condition in the furnace.

Figure 2. Pressure distribution along the furnace for various SA injection arrangements.
Temperature distribution along the furnace for different SA injection arrangements are demonstrated in Fig. 4. Temperature in the furnace firstly increases and then decreases in all experiments. The temperature in primary region is lower because of the adding of cold fuel and returning of the circulating materials at a relatively lower temperature. The temperature in upper region decreases due to the endotherm of water-cooling tube. The difference of the highest temperature and temperature in the upper region is within 30°C. The temperature distribution is relatively uniform in all experiments. The influence of injection of the third and fourth level SA on the temperature distribution is not significant.

From Fig. 5, it can be seen that the higher level SA injects, the lower NOx emission in the flue gas. As mentioned above, the reduction region is significantly lengthened in the furnace when higher SA injects, which can avoid much NOx produced from oxidation of nitrogen-containing precursors in the primary region and strengthen the NOx reduction reactions in the upper region, resulting in lower NOx emission. It also shows that NOx reduction is more obvious at higher temperature due to the intensified NOx reduction reactions at higher combustion temperature.

Another conclusion from Fig. 5 is that NOx emission decreases with combustion temperature increasing. Generally, NO emission increases with the increase in the combustion temperature, which has been verified by many researchers [4,6,7-8]. However, the trend is opposite for char. Related work has been done and can be seen in the future.
4. Conclusion
A CFB test rig is used to study the combustion and NO$_x$ emissions of char with higher secondary air (SA) injection. A good fluidized condition and uniform temperature distribution can be achieved with injection of higher level SA. NO$_x$ emission decreases with injection of higher level SA and the reduction effect is more obvious at higher temperature. NO$_x$ emission decreases with combustion temperature increasing for char combustion.

References
[1] J. M. Bursi, L. Lafanechere, L. Jestin. Basic design studies for a 600MWe CFB boiler (270b, 2*600°C), Proceedings of the 15th International Conference on Fluidized Bed Combustion, Savannah, Georgia, 1999.
[2] Zhen Fan, Archie Roberson, Steve Goidich. Foster Wheeler. 800MWe Circulating Fluidized Bed Boiler with 1300°F Supercritical Steam, 33rd International Technical Conference on Coal Utilization and Fuel Systems, Clearwater, Florida, USA, 2008.
[3] Arto Hotta, Kari Kauppinen, Ari Kettunen. Foster Wheeler. Towards New Milestones in CFB Boiler Technology-CFB 800 MWe/New 460 MWe Super-Critical Plant with CFB Boiler in Lagisza-First Experience Update, Power-Gen International 2010, Amsterdam, The Netherlands, 2010.
[4] L.F. de Diego, C.A. Londono, X.S. Wang, Bernard M. Gibbs. Influence of operating parameters on NO$_x$ and N$_2$O axial profiles in a circulating fluidized bed combustor, Fuel. 75 (1996) 971-978.
[5] Anders Lyngfelt, Lars-Erik Ámand, Lennart Gustavsson, Bo Leckner. Methods for reducing the emission of nitrous oxides from fluidized bed combustion, Energy conversion and management. 37 (1996) 1297-1302.
[6] A. Tourunen, J. Saastamoinen, H. Nevalainen. Experimental trends of NO in circulating fluidized bed combustion, Fuel. 88 (2009) 1333-1341.
[7] H. Kassman, M. Karlsson, Lars-Erik Ámand. Influence of air-staging on the concentration profiles of NH$_3$ and HCN in the combustion chamber of a CFB boiler burning coal, Fuel and Energy Abstracts. 41 (2000) 398.
[8] Carlos Lupiáñez, Luis I. Diez, Luis M. Romeo. Influence of gas-staging on pollutant emissions from fluidized bed oxy-firing, Chemical Engineering Journal. 256 (2014) 380-389.