Int. J. Therm. Sci. (2000) 39, 833–841

S1290-0729(00)00273-8/FLA

Comparison of nitrogen oxide emissions from boilers for a wide range of coal qualities

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(Received 24 September 1999, accepted 3 January 2000)

Abstract — This paper presents the results of experimental researches on nitrogen oxide emissions from coal-fired boilers. Two Chinese lean coals have been fired in two full scale boilers (1 025 $t\cdot h^{-1}$) and in a pilot scale test furnace (Drop-Tube Furnace) to study the influence of nitrogen content in coal on nitrogen oxide emission. The nitrogen oxide emission was found to correlate well with the fuel nitrogen content. The test results of the drop-tube furnace and the single burner furnace with three Chinese coals show that the staged combustion can greatly reduce the nitrogen oxide emission. Identical trends in nitrogen oxide emission as a function of the volatile matter of the coals have been obtained under different combustion conditions. The principle of low nitrogen oxide emissions of the wide range burner (biased combustion or internal air staging) and the cases with over fired air port (furnace air staging) is introduced in this paper. In addition, the influence of excess oxygen content on nitrogen oxide emission has been tested in the utility boilers and test furnaces. © 2000 Éditions scientifiques et médicales Elsevier SAS

coal quality / combustion experiment / nitrogen oxide

Nomenclature

ar	as received
CMR	continuous maximum ration
daf	dry, ash free
DC	direct current
DTF	drop-tube furnace
FZ1	Fuzhou power plant unit 1
HC3	Hanchuan power plant unit 3
LHV	low heating value
NO_x	nitrogen oxide
OFA	over fired air
QS12	Qingshan power plant unit 12
SBF	single burner furnace
WR	wide range
WF1	Wanfang power plant unit 1

1. INTRODUCTION

Today, a large amount of coals are utilized in power plants because of its huge resources in the world. One of the major concerns associated with coal-fired power plants is the emission of pollutants, especially nitrogen oxide. As we know, high levels of nitrogen oxide (NO_x) emitted into atmosphere may cause environmental pollution. Most countries, like China, have to bear the burden of environmental problems, and recently have specified limits for the emissions of NO_x. Moreover, because of the deregulation, the qualities of coals fired in boilers are changing frequently. The change of coal quality will have an impact on the performance of boilers and the NO_x formation. The operators of the utility boilers need a better knowledge to predict the change of performance in order to keep the emissions of NO_x below the country standard.

The influence of coal quality on furnace combustion performance has been investigated experimentally in recent years [1–15]. Some results concerning the influence of coal quality on NO_x formation have been found. These studies are almost conducted in pilot scale furnaces or drop tube furnaces. These results can only present the qualitative trends. *The influence of coal quality on combustion performance* both in full scale furnaces and in pilot scale furnaces has been investigated with considerable success, and a one-dimensional engineering model was developed [1]. In most of these experiments bitumi-

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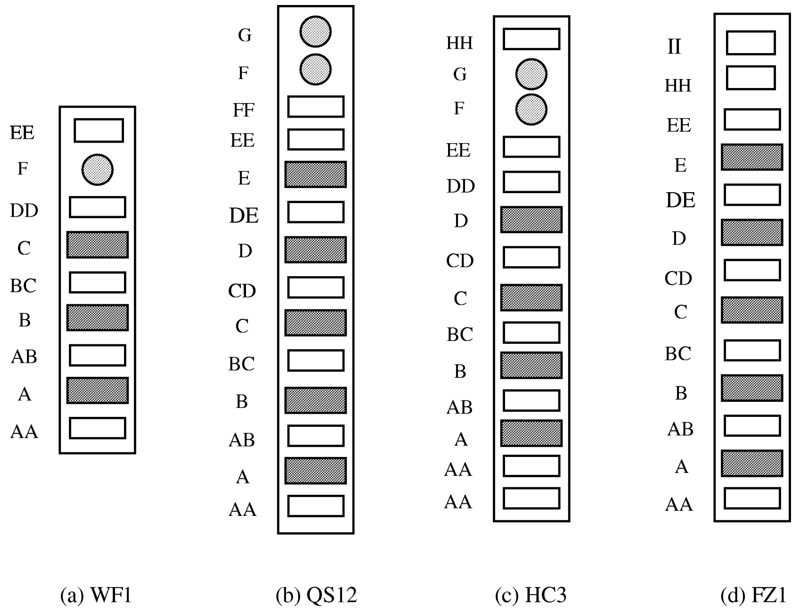

Figure 1. The burner arrangement. A, B, C, D, E: the primary air port; AA, AB, BC, CD, DE, DD, EE, FF: the secondary air port; F, G: the tertiary air port; HH, II: the OFA port.

nous coals have been fired. Although low NO_x burners and the over fired air (OFA) were introduced in the furnace design, the NO_x emissions of the boiler are higher when fired with anthracite or lean coal compared to bituminous coal. An experimental research program on low volatile content coals is of crucial significance for a wide range of coal qualities.

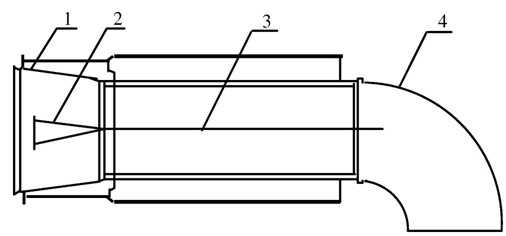

Figure 2. The structure of WR burner: 1. primary air nozzle, 2. corrugated bluff-body, 3. horizontal plate, 4. blend pipe.

2. EXPERIMENTAL FACILITIES AND PROCEDURE

2.1. Experimental facilities

The experiments have been performed in four full-scale boilers.

Wan Fang Power Station Unit 1 (WF1) is a 125 MWe corner fired boiler located in Henan Province, China. Anthracite is fired and the burners arrangement is shown in *figure 1(a)*.

Qing Shan Power Station Unit 12 (QS12) and *Han Chuan Power Station Unit 3* (HC3) are 300 MWe corner fired boilers located in Hubei Province, China. Lean coals are fired in both QS12 and HC3. The burners arrangement in QS12 is different from those in HC3, as shown in *figures 1(b)* and *(c)*. The OFA is added through 4 ports in one row above the burner belt in HC3.

Fu Zhou Power Station Unit 1 (FZ1) is a 350 MWe corner fired boiler located in Fujian Province, China. Bituminous coal is fired and the OFA is added through 8 ports in two rows above the burner belt, as shown in *figure 1(d)*.

A Wide Range (WR) burner is equipped in all of four utility boilers. *Figure 2* shows the structure of WR burner that has following characteristics. Firstly, the coal/air flow is centrifugally separated by a vertical bend pipe, and injected into furnace with dense as well as dilute coal/air jets eventually forming biased combustion. A relatively high concentration of pulverized coal in dense-phase air jet can ensure the ignition of coal particles. Biased combustion of dense and dilute phase will decrease

TABLE I
The features of four utility boilers.

	QS12	HC3	FZ1	WF1
Unit power (MW)	300	300	350	125
Boiler CMR ($t \cdot h^{-1}$)	1025	1025	1061.9	420
Coal type	lean coal	lean coal	bituminous coal	anthracite
Combustion type	tangentially fired	tangentially fired	tangentially fired	tangentially fired
Burner type	WR burner	WR burner	WR burner	WR burner
No. of the primary air ports (each corner)	5	4	5	3
No. of the secondary air ports (each corner)	7	7	6	5
No. of the tertiary air ports (each corner)	2	2	none	1
No. of the OFA ports (each corner)	none	1	2	None
Furnace dimension (width × depth × height)	14.08 × 11.858 × 53	11.76 × 11.97 × 49		9.6 × 8.84 × 36.2

NO_x formation. Secondly, a recirculating flow of gas will form after bluff-body near burner when coal/air jet is injected into furnace through corrugated bluff-body. This is beneficial to strengthen the contact of flue gas and the primary air flow, and enhance the heat and mass transfer. Thirdly, the circumference air decorated around the primary air nozzles can strengthen the rigidity of the primary air jet and adapt to the changing of coal type.

A brief description of the four utility boilers is given in *table I*.

2.1.1. Drop-tube furnace (DTF)

Figure 3 shows the schematic diagram of the drop-tube furnace. It is a 1 000 mm long refractory corundum tube with 40 mm internal diameter. The furnace is heated by electric heater of carborundum tube. The temperature in the furnace is measured by Pt–Rh thermocouple. The signal of the furnace temperature is connected with silicon controlled rectifier with responsibility for adjusting the power of electric heater. So the furnace can be a thermostat during the experiments. The external layer of the furnace is coated with thermal insulation material. The secondary air is heated by electric preheater and then blew into the furnace. The burner is located at the top of the furnace and fires downward.

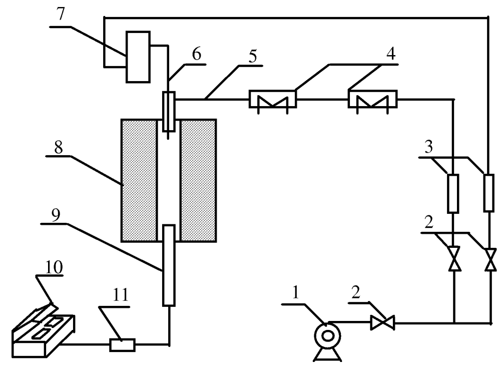

Figure 3. The schematic diagram of drop-tube furnace: 1. blower, 2. valve, 3. flowmeter, 4. air preheater, 5. secondary air, 6. primary air, 7. pulverized coal feeder, 8. furnace, 9. sample chamber, 10. gas analyzer, 11. filter.

2.1.2. Single burner furnace (SBF)

The furnace for the lab scale experiments in high temperature is 350 mm in height, 500 mm in width and 4 000 mm in length. The furnace with its 10 test holes of temperature and concentration and the air inlet ports is shown in *figure 4*. The coal feeder can provide pulverized coal with feeding rates in the range of 18–25 $kg \cdot h^{-1}$. The total combustion air is divided into four parts: the primary, below, middle and upper secondary air. The primary and secondary air can be preheated to 473 K and 573 K, respectively.

The pulverized coal mixed with the primary, below and middle air jets is ignited and fired in the early stage of the combustion process in the furnace. In general, this zone is named the main combustion region with stoichiometries ϕ_{MR} of 0.9 under staged combustion condition. Then the pulverized coal is mixed with the upper secondary air jet and burnout in the later stage of combustion process. This zone is named over fired region with stoichiometries $\phi_{MR} > 1.0$. The single burner furnace is able to simulate quantitatively the NO_x emissions of the utility boilers for the coals investigated.

2.2. Coal qualities

Four kinds of coal were tested in both the full scale furnaces and the pilot scale test rigs:

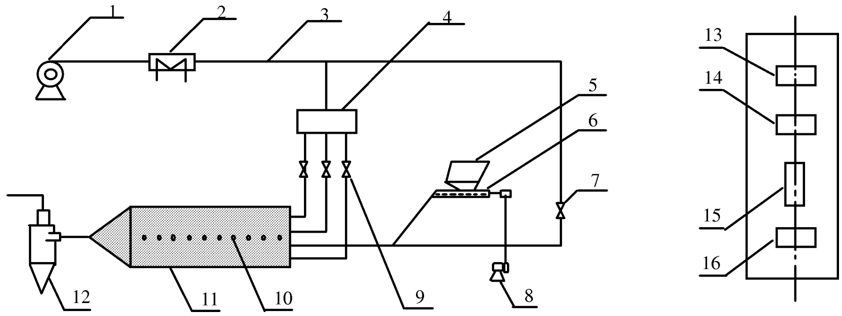

Figure 4. The schematic diagram of single burner pulverized coal furnace: 1. blower, 2. air preheater, 3. air pipe, 4. secondary air distributor, 5. pulverized coal hopper, 6. pulverized coal feeder, 7. primary air valve, 8. D.C. motor, 9. secondary air valve, 10. thermocouple, 11. combustion chamber, 12. cyclone separator, 13. upper secondary air port, 14. middle secondary air port, 15. primary air port, 16. below secondary air port.

TABLE II
Coal analysis (wt%) and size analysis.

	Coal 1			Coal 2			Coal 3				Coal 4		
	WF1	DTF	SBF	QS12	HC3	DTF	QS12	HC3	DTF	SBF	FZ1	DTF	SBF
C(daf)	89.4	89.4	90.0	91.5	92.4	91.5	89.4	90.5	89.4	89.4	82.92	84.44	84.44
N(daf)	1.50	1.50	1.40	1.88	1.90	1.88	1.42	1.44	1.42	1.42	1.12	1.41	1.41
Ash(ar)	12.06	12.06	11.34	18.1	18.6	18.1	27.7	28.2	27.7	27.7	19.77	25.73	25.73
Moisture(ar)	5.2	5.2	6.3	6.50	7.34	6.50	6.72	7.33	6.72	6.72	9.61	11.4	11.4
Volatile(daf)	9.80	9.80	9.73	17.1	16.6	17.1	19.1	17.6	19.1	19.1	32.31	31.68	31.68
LHV($KJ\cdot kg^{-1}$) (ar)	26 230	26 230	25 916	24 830	25 100	24 830	19 670	20 220	19 670	19 670	22 442	21 060	21 060
R_{90}	6.8	6.8	7.0	14	14	14	14	14	14	14	15*	18	18

* Here, $R_{74} = 15\,\%$.

- anthracite,
- a high nitrogen content and low ash content blending coal (lean coal),
- a low nitrogen content and high ash content blending coal (lean coal),
- bituminous coal.

The results of the coal analysis and size analysis are given in *table II*.

2.3. Test program

2.3.1. WF1, QS12, HC3 and FZ1

The test program in the utility boilers includes two configurations:

(1) *Table II* shows in detail the qualities of coals which were fired in the different utility boilers. Coal 2 and coal 3 were fired in QS12 and HC3 under the normal operation conditions (300 MWe, 4.2 % O_2 in the furnace outlet) to test the influence of coal quality on NO_x emission. The NO_x formation in WF1 for coal 1 was studied at full load (125 MWe) with 5.2 % O_2 at the furnace exit. Coal 4 was fired in the FZ1 at full load (350 MWe) with 2.5 % O_2 at the furnace exit to study the NO_x emission.

(2) In general, the amount of combustion air in furnace has an influence on NO_x formation. For the coal qualities specified in *table II*, the secondary air in four utility boilers was changed during the experiments in order to vary the amount of O_2 % at the furnace exit.

The experimental results of FZ1 boiler presented in this paper are referred from literature [5, 6].

During all the experiments, the concentrations of NO, NO_2 and O_2 were measured in the furnace outlet prior to the air preheater by gas analyzer.

2.3.2. DTF

All four coals were fired in the DTF to study the relationship of coal quality and NO_x formation, and to provide a common database for the comparison with the results from utility boilers. The temperature in combustion region of the furnace is about 1 473 K, and the particle residence time is around 0.5 s. The feeding rate of coals is 2 g·min^{-1}. The primary air ratio is about 10 % with 10 % O_2 at the furnace exit. The secondary air is heated to about 623 K by electric preheater. The operation condition of the DTF can ensure the burnout of the coals, and thermal-NO_x formation should not be taken into account when the temperatures of the furnace are lower than 1 300 °C. So the NO_x formation of the DTF is only fuel-NO_x, and this can compare the NO_x emissions with different coal qualities. The concentrations of NO, NO_2 and O_2 were measured at the furnace exit by gas analyzer.

2.3.3. SBF

Staged combustion tests were conducted in the SBF in order to investigate the influence of air staging on NO_x emission. *Table II* shows in detail the qualities of coals which were fired in the SBF. The pulverized coal was supplied to the burner with a feeding rate of 22 kg·h^{-1}. Under well controlled conditions, for a reliable transport of the pulverized coal the primary air ratio and the below secondary air ratio of the furnace were maintained at its minimum value (roughly 30 %). The middle secondary air ratio and the upper secondary air ratio were changed with main combustion region stoichiometries ϕ_{MR} of 0.9 and 1.0, and 4.0 % O_2 at the furnace exit.

During all the measurements, the gas temperatures along burner centerline are measured by water-cooled Pt–Rh thermocouples. The thermocouples were calibrated against a suction pyrometer. A water-cooled sampling probe is used to extract gaseous products of combustion for gas analysis (O_2, NO and NO_2).

3. RESULTS AND DISCUSSION

3.1. The influence of fuel bound nitrogen

Figure 5 shows the NO_x emission as a function of the coal quality in three furnaces. The data show that combustion of coal 2 results in a higher amount of NO_x followed by coal 3 in QS12 and HC3. The difference of the NO_x emissions of coal 2 and coal 3 can be explained by the higher nitrogen content of coal 2. The coal nitrogen is evolved and oxidized to NO_x when it is mixed with the secondary air. It is reasonable that a larger fraction of the coal nitrogen

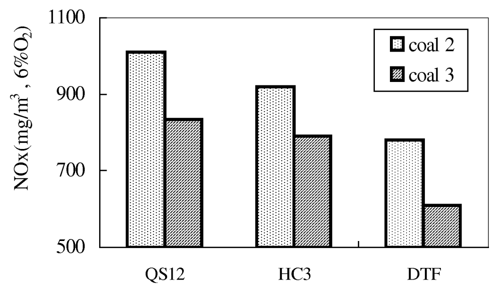

Figure 5. The relationship of coal type and NO_x emission. QS12: 300 MW, 4.2 % O_2; HC3: 300 MW, 4.2 % O_2; DTF: $T =$ 1 473 K, 10 % O_2.

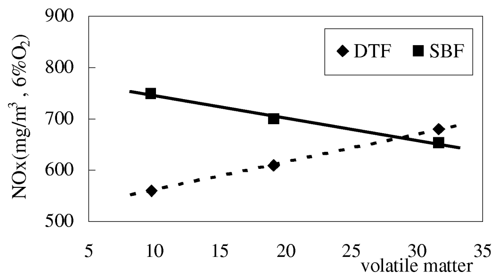

Figure 6. The relationship of volatile and NO_x formation. DTF: single-stage, $T = 1\,473$ K, 10 % O_2 at the furnace exit. SBF: two-stage, the main region stoichiometric ratio is 0.9 and 4.0 % O_2 at the furnace exit.

results in higher NO_x emissions. An increase from 1.4 wt% coal-N to 1.9 wt% resulted in about 150 mg·m^{-3} more NO_x being produced. Good agreement between the NO concentrations and the coal-N content has been previously obtained by Pieter [1] and Afonso [2].

Because of the lower temperature (1 473 K), thermal-NO_x formation should not be taken into account. The NO_x formation of the DTF is only fuel-NO_x. Moreover, because of shorter residence time (around 0.5 s), the NO_x formation in the DTF may further decrease. Although the quantitative agreement is not more than satisfactory, both the power stations and the pilot scale test rigs show the same qualitative trends in the correlation of the NO_x concentrations with the coal nitrogen content.

3.2. The influence of volatile matter

We can see the influence of volatile matter on the NO_x formation of the SBF and the DTF in *figure 6*. Increasing the volatile content of the coal results in increasing nitrogen oxide emissions under single stage combustion conditions but decreasing emissions of nitrogen oxide under air staged combustion conditions. This result is consistent with literature [2].

NO formation model [7, 8]:

$$\text{decomposition} \quad \text{coal-N} \rightarrow HCN + \cdots \tag{1}$$

$$\text{oxidation} \quad HCN + O_2 \rightarrow NO + \cdots \tag{2}$$

$$\text{reduction} \quad HCN + NO \rightarrow N_2 + \cdots \tag{3}$$

When coals with higher volatile content were fired under staged combustion condition, the coal-N may decomposes to more intermediate products of HN, HCN, CN and NH. As a consequence, the reduction of NO to N_2 (reaction equation (3)) in the fuel-rich combustion region is augmented. The influence of the staged combustion on the NO_x formation was investigated experimentally in the SBF.

In the DTF, because coals were fired under oxygen-rich condition, the coal-N decomposes to HCN, and is then converted to a large extent of NO. For that reason, it is easy to understand that high volatile bituminous coals may produce more NO_x than low volatile anthracite under single stage combustion conditions.

3.3. The influence of the amount of excess air

Figure 7 shows the relationship of the O_2 levels with the NO_x concentration. The data show that NO_x emissions increase initially and then decrease slightly with the increase of the amount of excess oxygen. The phenomenon may be that a higher oxygen concentration may provide oxidizing conditions for coal nitrogen transforming to fuel-NO_x. In addition, sufficient oxygen may enhance the combustion intensity and result in a high flame and furnace temperature. Locally high temperature may slightly increase thermal-NO_x formation according to Zeldovich's mechanism [16]. If the oxygen content is further increased, the furnace temperature on the main combustion region may decrease. This may restrain the combustion intensity and the combustion rate and may result in a low amount of NO_x formation. For that reason, the upward trend may be reversed with the increase of the amount of excess air, after a certain threshold limit is exceeded.

3.4. The influence of the air staged combustion

Figure 8 shows the NO_x emissions of three coals fired in the SBF with the main region stoichiometries ϕ_{MR} of 0.9 and 1.0, and the measured temperature distribution of the furnace along the burner center are shown in *figures 9–11*. It can be seen that NO_x formation may decrease when the middle secondary air ratio was decreased. Because of the staged combustion ($\phi_{MR} = 0.9$), the oxygen concentrations may decrease in the early stage of the combustion process. This makes the main combustion region fuel-rich. As a consequence, this leads to a lower combustion intensity and decreases the peak value of temperature (see *figures 9–11*) in main combustion region. The coal-N will decompose to HN, HCN, CN and NH and the NO conversion to N_2 is improved in the fuel-rich combustion region. In sum, the staged combustion may decrease NO_x formation, especially for highly volatile coals.

Figure 12 shows that the characteristic of the NO_x formation in the HC3 boiler is superior to that in the QS12 boiler. Comparing the burner structure and arrangement of the two units, it must be concluded that the main contribution to the smaller NO_x production of HC3 boiler comes from the furnace air staging with OFA. The influence of OFA on the NO_x formation has been also found in the previous works [15, 17–19]. The schematic

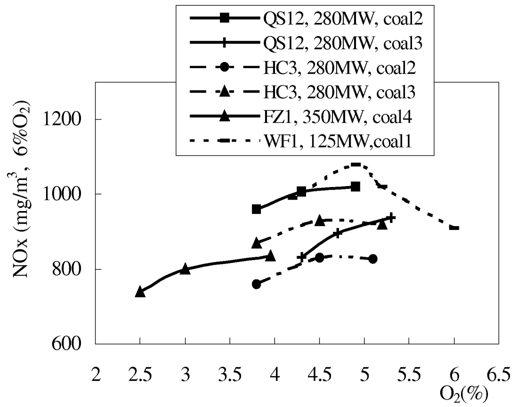

Figure 7. The relationship of O_2 levels and NO_x emission.

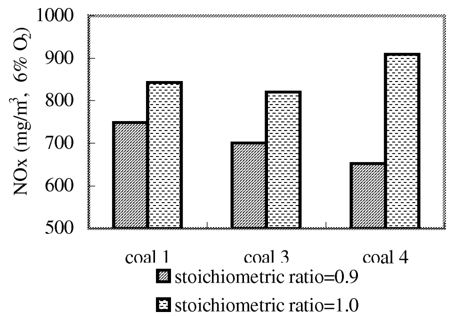

Figure 8. The influence of air staging on the NO_x emission.

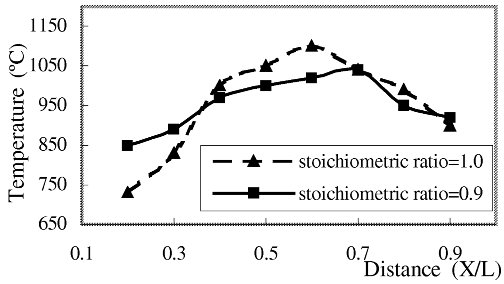

Figure 9. Furnace temperature distribution along the burner centre (coal 1 fired in SBF).

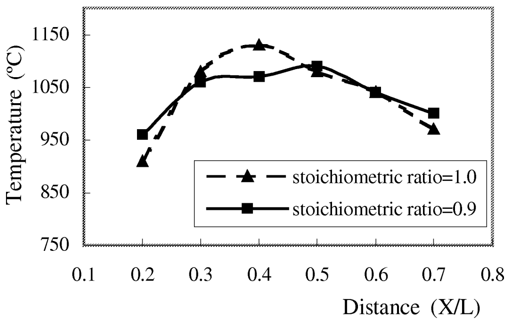

Figure 10. Furnace temperature distribution along the burner centre (coal 3 fired in SBF).

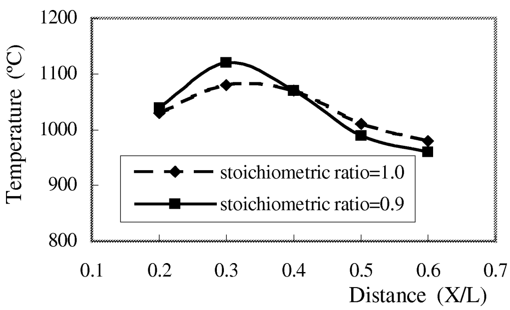

Figure 11. Furnace temperature distribution along the burner centre (coal 4 fired in SBF).

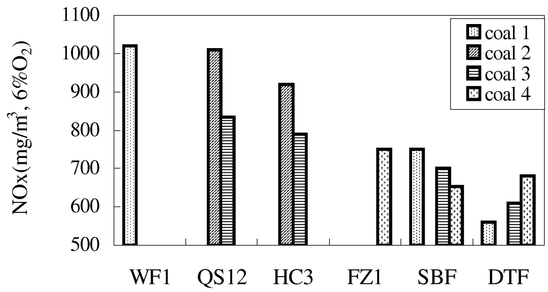

Figure 12. The NO_x emission from different furnaces with different coals. WF1: 125 MW, 5.2% O_2; QS12: 300 MW, 4.2% O_2; HC3: 300 MW, 4.2% O_2; FZ1: 350 MW, 2.5% O_2; SBF: 4% O_2; DTF: $T = 1473$ K, 10% O_2.

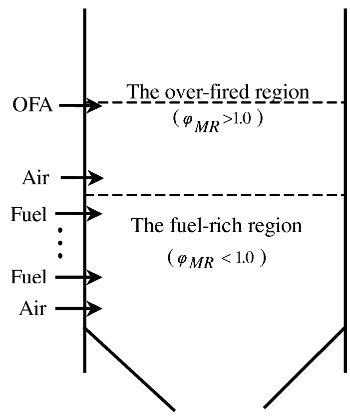

Figure 13. The schematic diagram of the furnace air staging with the OFA in utility boilers.

diagram of furnace air staging with the OFA in utility boilers is shown in *figure 13*. As aforementioned principle, for the staged combustion the main combustion region is fuel-rich with stoichiometries $\phi_{MR} < 1.0$. Here the volatile nitrogen and char nitrogen is evolved to intermediate products in the reducing atmosphere. As a result, reduction reaction of the NO conversion to N_2 is enhanced. This certainly leads to the lower generation of NO_x. The coals can usually be burned completely in the over-fired region. The FZ1 boiler emitted the lowest NO_x emissions within four utility boilers because of the high volatile matter content of the coal fired and the furnace air staging with OFA. Because ignition stability and burnout performance of pulverized coal must be considered in boiler design and operation for low volatile matter anthracite, the technology of the furnace air staging with OFA cannot be introduced in the WF1 boiler. Compared to the air staging, single stage combustion decreases the residence time of the pulverized coal particles in a reducing environment, and leads to the highest NO_x production of the WF1 boiler.

The key technologies for a low NO_x formation are the position of OFA ports (the distance to upper secondary air port) and the OFA ratio when the furnace air staging with the OFA is introduced in the boiler. In the experiments with the FZ1 and the HC3 boilers the OFA ratio (the proportion of the OFA flow to the total secondary air flow) was about 11.5% and 8%, respectively. This is not enough to make the early stage of combustion under stoichiometric air/fuel ratio of coal burnout, which explains that the NO_x emissions in the HC3 and the FZ1 furnace were still high, although low NO_x combustion technologies have been adopted.

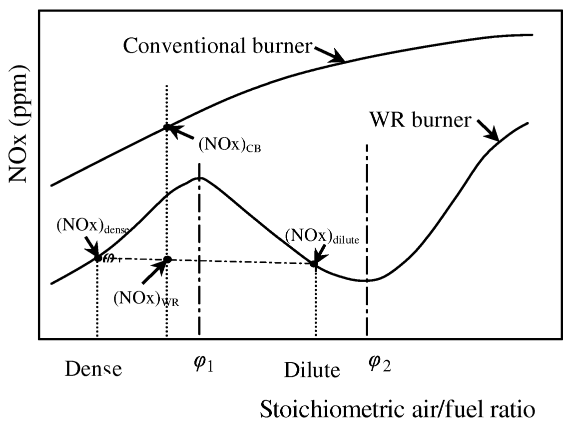

Figure 14. The principle of NO_x reduction with the WR burner. φ_1: stoichiometric ratio of volatile combustion, φ_2: stoichiometric ratio of coal burnout.

Another staged combustion named biased combustion with WR burner is adopted in the four utility boilers. The combustion performance of the WR burner has been investigated in the past [20]. A WR burner improves performance of ignition, stabilizes combustion and reduces NO_x formation. Contrast with furnace air staging with OFA, biased combustion with the WR burner is named internal air staging combustion, and the principle of NO_x reduction with the WR burner is shown in *figure 14*. In this figure $(NO_x)_{dense}$ represents the NO_x production of the dense-phase flame, and $(NO_x)_{dilute}$ represents the NO_x production of the dilute-phase flame. The overall NO_x production of WR burner is the average value $(NO_x)_{WR}$ of $(NO_x)_{dense}$ and $(NO_x)_{dilute}$. Compared to the $(NO_x)_{CB}$ of conventional burner, the NO_x formations of WR burner greatly decrease. Although the quantitative decrease of NO_x formation cannot be given because all the utility boilers are equipped with WR burners, it is confirmed that the NO_x emissions from four utility boilers with the WR burner are lower than those from the boilers with the conventional burner. Moreover, further studies on the technologies of low NO_x emission will be necessary, especially for boilers fired with lean coal and anthracite.

4. CONCLUSION

From the experimental research conducted in this paper, the following conclusions can be made:

- The coal quality may affect remarkably NO_x emissions from boilers. An increase from 1.4 wt% coal-N to 1.9 wt% results in about 150 $mg \cdot m^{-3}$ more NO_x being produced.
- The emissions of nitrogen oxide are influenced by the volatile matter of the coals. An increase of the volatile content of the coal results in an increase of the nitrogen oxide emissions under single stage combustion conditions but leads to decrease of the emissions under air staged combustion conditions.
- The staged combustion may reduce the NO_x emissions significantly, especially for highly volatile coals. A WR burner may improve the performance of ignition, stabilize combustion and reduce NO_x emissions. The lower NO_x production of the HC3 boiler compared with the QS12 boiler comes from furnace air staging with OFA. The NO_x formation of the FZ1 boiler is the lowest within four boilers. This is caused by the highly volatile bituminous coal fired and the staged combustion with the OFA.
- Because ignition stability and burnout performance of pulverized coal must be considered in the boiler design and operation, combustion intensity must be increased for low volatile matter anthracite. This makes the WF1 boiler produce the highest NO_x emissions of four utility boilers.
- The experiments with the SBF show that the increase of upper secondary air (OFA) ratio can reduce remarkably the NO_x formation.
- Because the OFA ratio is too low, the NO_x emissions in the HC3 and the FZ1 furnaces are still high, although low NO_x combustion technologies have been introduced. Choosing a proper OFA ratio is the key technology to ensure combustion stability, high combustion efficiency and low pollution. Moreover, further studies on technologies of low NO_x emission will be necessary especially for boilers fired with lean coal and anthracite.
- The tests of the NO_x formation of the full scale boilers show that NO_x emissions increase initially and then decrease slightly when the oxygen content of the furnace exit is increased.

Acknowledgements

The experiments in Qingshan, Hanchuan and Wanfang power plants were performed with support from Li Min, Zou Zuqiao, Cheng Xiaohui, Han Dongmao, Liu Shengrong and Zuo Zhixiong. The experiments of single burner furnace were operated by Xiao Jingfang. Coal analysis was conducted by Wang Hong. All the laboratory experiments were conducted in the National Laboratory of Coal Combustion, Huazhong University of Science and Technology.

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