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NO$_x$ EMISSIONS FROM BUBBLING FLUIDIZED BED COMBUSTION OF LIGNITE COAL

Pavel Skopec*, Jan Hrdlička, Jan Opatřil, Jiří Štefanica

Department of Energy Engineering, Faculty of Mechanical Engineering, CTU in Prague, Technická 4, Praha 166 07, Czech Republic

* corresponding author: `p.skopec@fs.cvut.cz`

Abstract. This paper presents experimental results of NO$_x$ emission measurements for combustion of two kinds of coal in a bubbling fluidized bed combustor. The tested fuels were Czech brown coal (CBC) and German young brown coal (GYC). These fuels have different nitrogen contents. The experiments were performed in the pilot scale BFB experimental unit with power output of 500 kW. The influence of several parameters on NO$_x$ formation are investigated in this paper. The parameters studied here include the effect of the nitrogen content in the fuel, the effect of the oxygen concentration in the bed, the effect of bed temperature, the effect of air staging, and the effect of fluidization velocity. Significantly different behaviour of the fuels was found. Although GYC has a lower nitrogen content than CBC, it is more reactive and produces higher NO$_x$ emissions. The biggest dependence of NO$_x$ production for CBC was found for the effects of air staging and fluidization velocity. As the fluidization velocity increases and the amount of secondary air decreases, there is an increase in NO$_x$ emissions. The oxygen concentration in the bed has the strongest effect on the NO$_x$ production of GYCs. With increasing oxygen concentration, the production of NO$_x$ also increases. On the basis of the NO$_x$ measurements, the N-NO conversion factor was calculated and the effect of the operating parameters on this conversion factor was investigated.

Keywords: NO$_x$ emissions; coal combustion; fluidized bed.

1. Introduction

Fossil fuels play a crucial role in the energy mix, and will continue to play a major role in decades to come. Coal is the most common source for heat and power production, and the role of coal will continue to be very important in the near future. According to EIA statistics for 2013, world coal consumption was 8.1 billion tonnes, of which more than 75 % was used for electricity production [1]. Nitrogen oxides, which are released during coal combustion, are considered as major pollutants. The most important nitrogen oxides are NO and NO$_2$, and NO$_x$ is used to refer to the total amount of nitrogen oxides. About 95 % of nitrogen oxides from industrial activities come from combustion processes, both from stationary combustion sources (about 40 %) and from mobile combustion appliances, i.e., motor vehicles (about 60 %). About 22.5 % of NO$_x$ is assigned to energy generation and distribution, while 13 % is assigned to industrial energy. About 13 % of NO$_x$ is produced in households [2]. The percentage of NO in the whole sum of NO$_x$ is much higher (about 90–99 %) than the percentage of NO$_2$ (about 1–10 %) [3]. Nitrogen oxides have a strongly negative environmental impact. In the stratosphere they convert to nitric acid, which causes acid rains. In addition, NO acts to deplete the stratospheric ozone layer. NO and NO$_2$ are toxic, and can cause respiratory problems. Another problem is photochemical smog. It arises as a reaction of UV rays with nitrogen oxides in the atmosphere, producing ground-level ozone.

2. NO$_x$ emissions from fluidized bed combustion

In general, three NO$_x$ formation mechanisms have been described:

- thermal mechanism — NO$_x$ formation from atmospheric nitrogen N$_2$;
- fuel mechanism — NO$_x$ formation from nitrogen compounds present in the fuel;
- prompt mechanism — NO$_x$ formation from nitrogen in the air and from hydrocarbon radicals.

Thermal NO$_x$ is formed at high temperatures by a radical reaction of N$_2$ and oxygen radical from the combustion air. These reactions were proposed by Zeldovich [4]. The main factors that affect the formation of thermal NO$_x$ are the maximum temperature and concentration of atomic oxygen in places with high temperatures. The thermal route of NO$_x$ formation becomes significant at 1200 °C, and rises exponentially with temperature. Fluidized bed combustion is characterized by lower operation temperatures, typically below 950 °C. For this reason, thermal NO$_x$ does not affect final amount of NO$_x$ emissions. Prompt NO$_x$ (also called Fenimore NO$_x$) is formed by the reaction of air nitrogen radicals with hydrocarbon radicals on the flame boundary, and it is most prevalent in rich flames. The process is characterized by several factors — the reaction occurs very rapidly, the temperatures are above 1500°C, and there is high dependency on the excess air with the maximum around the stoichio-

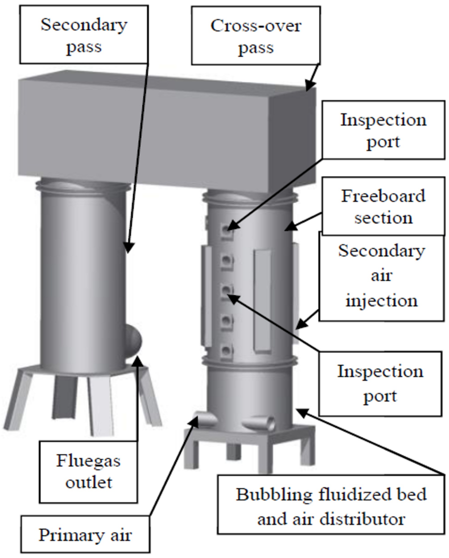

Figure 1. Basic scheme of the experimental BFB boiler.

metric ratio [4]. The formation of prompt NO_x starts with reaction

$$CH + N_2 \rightarrow HCN + N. \qquad (1)$$

The amount of prompt NO_x in fluidized bed combustion is negligible.

Fuel NO_x is the main source of NO_x production in FBC. It is formed by oxidation of the nitrogen contained in a fuel. One pathway is via evaporation of combustible matter; the fuel nitrogen that is released in the volatiles consists mostly of HCN and NH3. These products further oxidize to NO by reacting with OH radicals. Another way is via production from char nitrogen, which remains chemically bonded in aromatic structures to the char after devolatilization. The amount of NO_x that is produced depends strongly on the nitrogen content in the fuel. However, the correlation between N content in the fuel and NO_x production is not proportional. Only a part of the N is converted to NO. [5]

The ratio between nitrogen that is converted into flue gas in the form of NO and the total amount of nitrogen in the fuel is called the conversion factor, and is defined by the following equation:

$$\frac{\text{N in the fuel converted to NO}}{\text{organically bounded N in the fuel}} < 1. \qquad (2)$$

The main factors influencing the production of fuel NO_x are the amount of nitrogen content, the concentration of molecular oxygen in the zone of burning, and the temperature. This paper reports on a study of the effects of these factors on the production of NO_x emissions during fluidized bed combustion of lignite coal. The investigated parameters are:

- the effect of different nitrogen contents in the coal — two different types of fuels are investigated;
- the effect of the concentration of molecular oxygen in the zone of burning, two factors are investigated — the effect of excess air and the ratio between primary and secondary air;
- the effect of fluidization velocity;
- the effect of bed temperature.

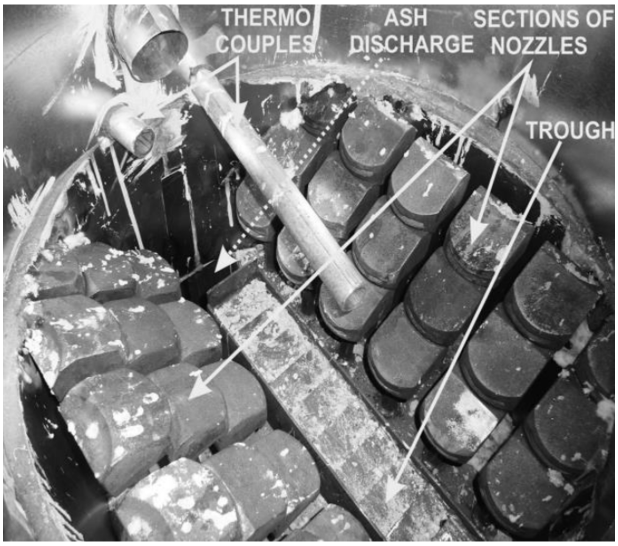

Figure 2. Fluidized bed distributor.

3. Experimental procedure

The experiments were performed on an experimental bubbling fluidized bed boiler with power output around 500 kW. The boiler is designed as a vertical double pass boiler with a circular cross-section having a cross-over pass that connects the two vertical passes. The basic scheme is shown in Fig. 1. The boiler is equipped with a V-shaped trough type of fluidized air distributor, and consists of 36 nozzles immersed in the fluidized bed, which are located on two parallel sides and are placed horizontally in three cascade rows (see Fig. 2).

The first pass of the boiler, including the fluidized bed distributor, is designed as an almost adiabatic combustion chamber. The chamber has a firebrick lining, with a water cooling double wall on the outer side. The cross-over chamber is also cooled by water walls. A secondary pass is made as a fire tube heat exchanger. Fluidization air is supplied by a primary fan with controllable revolutions. Secondary air is supplied via a separate ventilator with the possibility of controllable revolutions. Secondary air is supplied to a special distributor, from which it is led to the freeboard in four high levels and four places around the circumference of the first pass. Secondary air can therefore be added at 16 points, and each point is provided with a flap to enable better regulation. The boiler is also equipped with recirculation of the flue gas. The flue gas is recirculated in the place behind the cyclone to dust the flue gas off, and is supplied to the primary air duct. In the case of an adiabatic

Fuel	C^r [%]	H^r [%]	N^r [%]	S^r [%]	A^r [%]	W^r [%]	V^{daf} [%]	$Q_i{}^r$ [MJ/kg]
CBC	23.37	2.05	0.47	1.58	36.94	25.65	53.9	8.72
GYC	28.78	2.53	0.35	1.09	13.26	43.61	57.9	10.54

Table 1. Analysis of the fuel samples.

	Correlation index	
Parameter	**CBC**	**GYC**
Concentration of oxygen in flue gas	0.59	0.73
Primary/secondary air flow	0.85	0.45
Fluidization velocity	0.76	0.17
Bed temperature	−0.18	−0.18

Table 2. Results of a correlation analysis of NO$_x$ emissions and tested parameters.

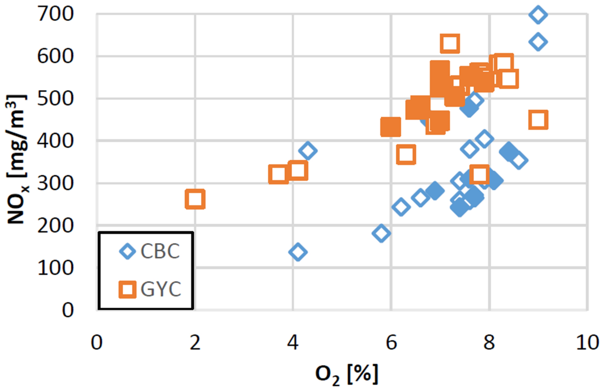

Figure 3. Effect of oxygen concentration on NO$_x$ emissions.

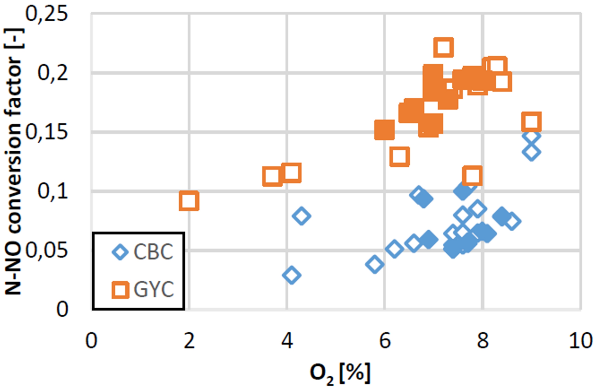

Figure 4. Effect of oxygen concentration on the N–NO conversion factor.

combustion chamber, recirculation of the flue gas is an effective way of regulating the fluidized bed temperature. Fuel is loaded into the boiler by a screw conveyor, which brings the fuel from the container located next to the boiler. The power capacity of the boiler is controlled by adjusting the fuel loading interval. The sampling point for online emissions analysis is located in the exhaust tube just behind the second pass of the boiler. The measured emissions were CO, O_2, SO_2, TOC (total organic carbon) and NO$_x$. The fluidized bed temperature was measured at two points, and the mean value was finally used for further evaluation. The amount of primary and secondary air were measured using thermo-anemometers placed in the primary and secondary air supply.

Two kinds of coals were used as the fuels for the experiments — Czech brown coal (CBC) from the North Bohemian coal basin, and German young brown coal (GYC) from Leipzig, in central Germany. Table 1 presents the results of a proximate and ultimate analysis of the coals.

As can be seen, both fuels are of low quality, with a relatively low heating value. CBC typically has a high ash content, while GYC typically has a high water content and a relatively low ash content.

4. Results and discussion

The combustion experiments were carried out in such a way that all parameters except one were kept constant for each experiment. This variable parameter was changed within a selected range. Each experiment lasted at least one hour under stabilized conditions, and the operation parameters of the boiler and the emission levels in the flue gas were continually recorded. The data on NO$_x$ concentrations in volume ppm were converted to mass concentrations related to dry flue gas at normal pressure and temperature and a reference oxygen content of 6 %. The following parameters were investigated:

- O_2 concentration in the bed in the range from 2 to 8 %
- fluidization velocity in the range from 0.5 to 1.2 m/s
- bed temperature from 790 to 860 °C
- primary and secondary air ratio from 0.5 to 7.

Each of the tested parameters can affect the formation of NO$_x$. On the basis of the measured data, a correlation analysis with 95 % reliability was performed. The results are summarized in Tab. 2. The table contains correlation indexes be-

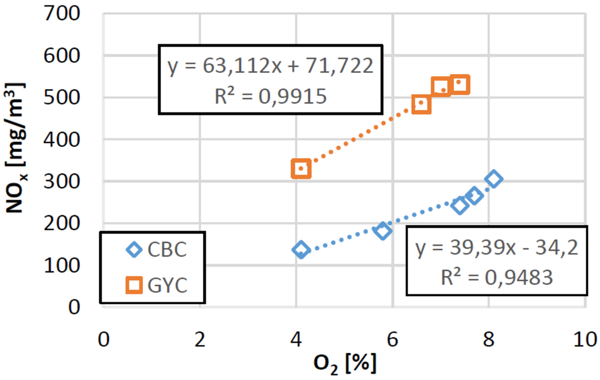

Figure 5. Effect of oxygen concentration on NO_x production for selected data.

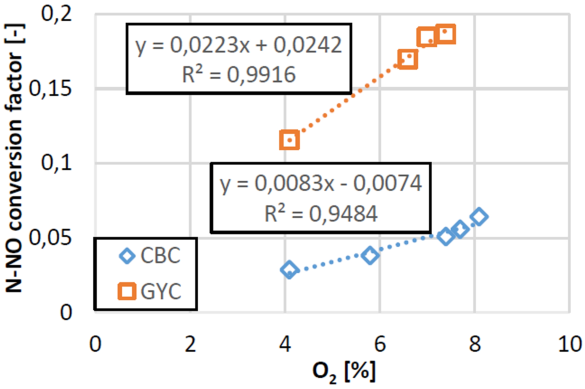

Figure 6. Effect of oxygen concentration on N–NO conversion factor for selected data.

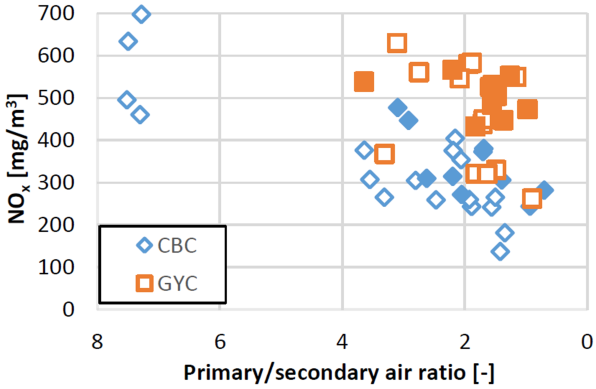

Figure 7. Effect of primary/secondary air ratio on NO_x emissions.

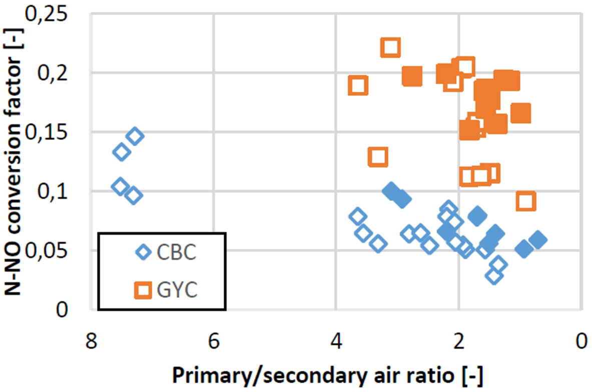

Figure 8. Effect of the primary/secondary air ratio on the N–NO conversion factor.

tween the level of NO_x and a particular parameter. The data in Tab. 2 shows the different behaviour of the fuels. The strongest dependence of NO_x formation for CBC is on the ratio between primary air flow and secondary air flow, and on the fluidization velocity, which is somewhat weaker. The effect of the concentration of oxygen in the flue gas with correlation index 0.59 also shows significant dependence. GYC shows completely different behaviour. The concentration of oxygen in the bed has the strongest effect on NO_x emissions. Air staging also has some effect, but there is a significantly lower correlation index than for CBC. The effect of temperature is very low for both fuels.

4.1. Effect of oxygen concentration

The effect of oxygen concentration on the NO_x emissions is shown in Fig. 3. The chart also includes data from measurements with higher limestone addition (marked with filled points). It can be observed that greater addition of limestone (from Ca/S 1.5 to 3) does not affect NO_x production. We can see that with increasing oxygen concentration the production of NO_x also increases. Similar behaviour was identified in Fig. 4 with the effect of oxygen concentration on the N–NO conversion factor.

The combined effect of oxygen and air ratio can make it difficult to distinguish between these two effects. In order to separate off the effect of oxygen concentration, it was necessary to select the data appropriately. The selection was based on choosing experimental data with the same primary/secondary air ratio. Variability of the primary/secondary air ratio up to 5 % was considered acceptable. The range of the ratio was between 1.35 and 1.56 for CBC, and between 1.49 and 1.59 for GYC. The results for selected data are shown in Fig. 5 and Fig. 6. The values were fitted by linear regression with a determination index higher than 0.9 for all cases. Similar results were also found for other data selection intervals.

Our findings indicate that increasing oxygen concentration has an increasing effect on NO_x production. This corresponds to findings reported in the literature [3, 5], with the explanation that a rising oxygen concentration increases the combustion rate. As a result, homogeneous reactions for the oxidation of HCN and NH3 are enhanced, while heterogeneous reactions for the reduction of NO are supressed [5].

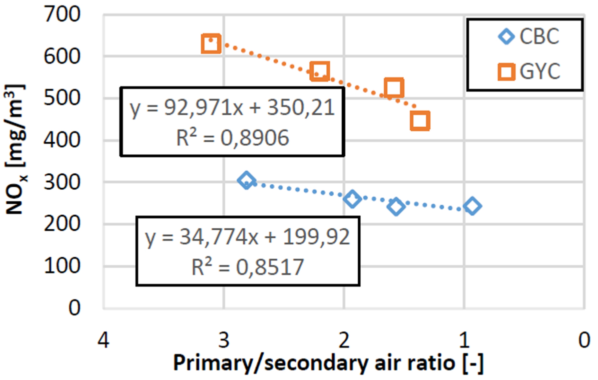

Figure 9. Effect of the primary/secondary air ratio for selected data on NO$_x$ emissions.

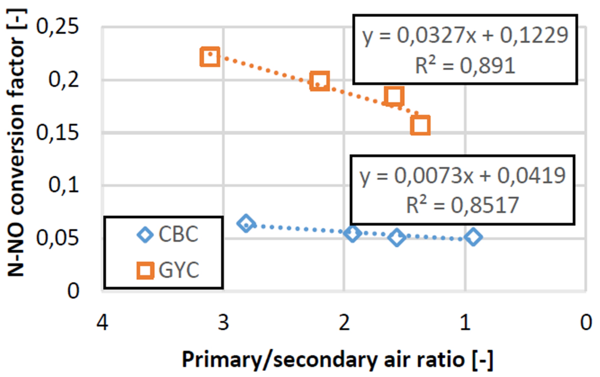

Figure 10. Effect of the primary/secondary air ratio for selected data on the N–NO conversion factor.

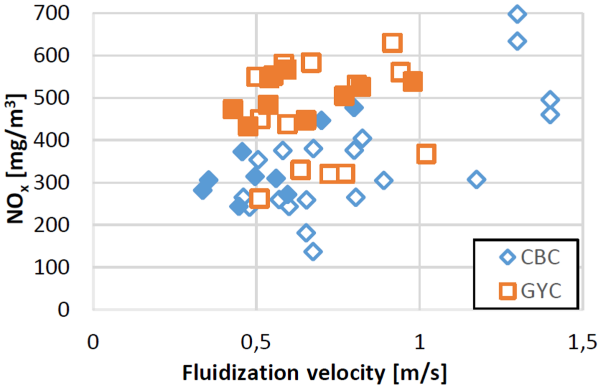

Figure 11. Effect of the fluidization velocity on NO$_x$ emissions.

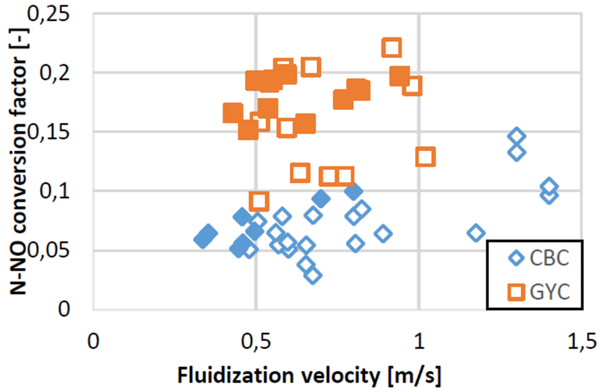

Figure 12. Effect of the fluidization velocity on the N–NO conversion factor.

4.2. Effect of air staging

The effect of air staging is defined by the ratio between the primary air flow and the secondary air flow. Values of NO$_x$ emissions in dependence on this ratio from all experiments are shown in Fig. 7 and Fig. 8. We can see that the NO$_x$ emissions for GYC show no clear dependence on the primary/secondary air ratio, and there is no clear trend, but that CBC shows an increasing trend. This also corresponds with the correlation analysis, which gives greater weight to the air ratio for CBC.

A similar approach for data selection as in the previous case was used here in order to eliminate the combined effect with oxygen concentration. Selected experiments found a similar oxygen concentration of 11.4 % for CBC and 11–11.2 % for GYC. The results are shown in Figs. 9 and 10. The values were again fitted by linear regression, and the determination index was above 85 % for all cases. Similar results were also found for other data selection intervals.

Figures 9 and 10 show that an increase in the primary to secondary air ratio, i.e. a decrease in the secondary air flow, causes an increase in NO$_x$ emissions. GYC shows steeper dependence than CBC. The same observations are also valid for the N-NO conversion factors. The decrease in NO$_x$ emissions with increasing secondary air flow (a decreasing primary/secondary air ratio) can be attributed to an enhanced reducing atmosphere in the bed. Under staged combustion, the char and CO concentrations in the bed increase, therefore enhancing the rates of NO reduction on the char and, in addition, more fuel N is decomposed as N_2 [5].

4.3. Effect of fluidization velocity

The effect of fluidization velocity on NO$_x$ formation is shown in Figs. 11 and 12. We can observe a similar effect to the ratio between primary and secondary air. For GYC, there is no clear dependence of these two parameters, but CBC has a slightly increasing character. This also corresponds with the correlation analysis, which shows higher dependence for CBC (correlation index 0.76) than dependence for GYC (correlation index 0.17).

Data selection was used in a similar way here, in order to eliminate unwanted combined effects of oxygen content. The results are shown in Figs. 13 and 14. The selected experiments have a similar oxygen concentration as in the previous example (11.4 % for CBC

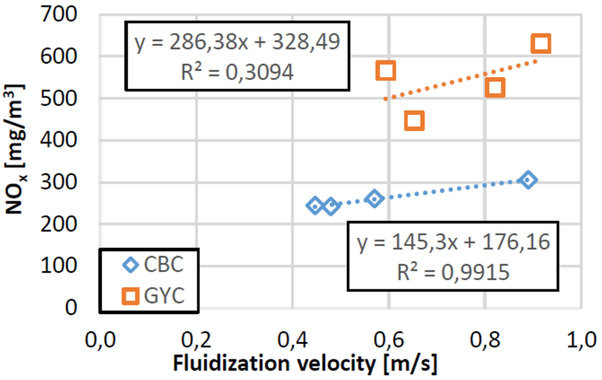

FIGURE 13. Effect of the fluidization velocity for selected data on NO_x emissions.

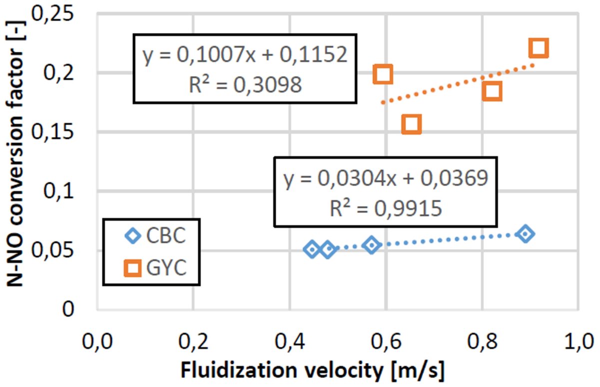

FIGURE 14. Effect of the fluidization velocity for selected data on the N–NO conversion factor.

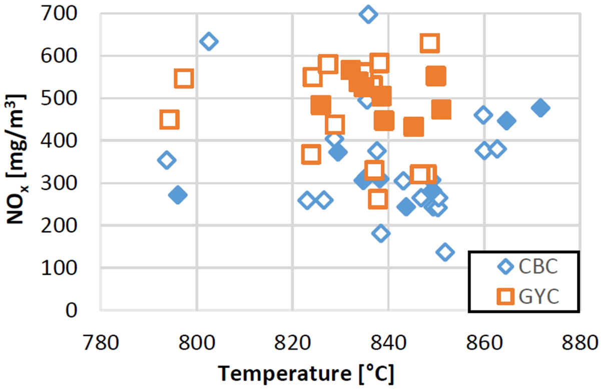

FIGURE 15. Effect of bed temperature on NO_x emissions.

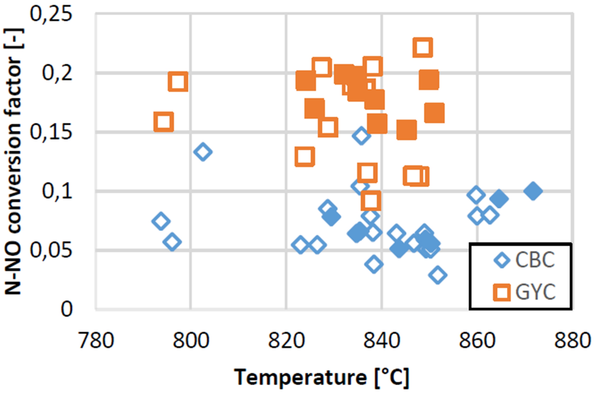

FIGURE 16. Effect of bed temperature on the N–NO conversion factor.

and 11–11.2 % for GYC). Almost linear dependence can be observed for CBC, but there is no specific dependence for GYC.

4.4. EFFECT OF BED TEMPERATURE

The bed temperature was changed from 790 to 870 °C, which corresponds to the general operating conditions in most FBCs. No clear dependence of NO_x production on fluidized bed temperature was found. This was confirmed by correlation analysis (the lowest correlation index in Tab. 2), and also by Figures 15 and 16. A literature search produced no clear results. Most studies in the literature support the general statement that an increase in bed temperature leads to an increase in NO_x production [5]. This is explained by the fact that an increase in the combustion temperature increases both the combustion rate and the concentration of free reactive radicals. At the same time, higher temperatures promote the oxidation of nitrogen radicals. A higher combustion rate reduces the production of char and CO. This in turn decreases the heterogeneous reduction of NO to N_2 on the char surface. However, a small number of results are in contradiction with this general trend of NO emissions [5]. A possible explanation is that an increase in bed temperature promotes fuel devolatilization in the bottom dense zone, where volatile-N is mainly oxidised to N_2. As a result, less char-N is carried over to the oxygen-rich zone, where it is oxidised to NO [5].

4.5. EFFECT OF NITROGEN CONTENT

The effect of nitrogen content was studied by using two different coals with two different nitrogen contents (CBC 1.3 % N in dry-ash free basis; GYC 0.8 % N in dry-ash free basis). The results show that although GYC has a lower N content, it has an almost twofold higher N–NO conversion factor. This is in contradiction with the general statement that an increase in nitrogen content leads to increased NO_x production. The degree of nitrogen conversion depends on the fuel reactivity and the fuel characteristics. Some role is also played by the fuel-volatile content — an increase in fuel-volatile content leads to an increase in NO_x.

5. CONCLUSIONS

Several operating parameters and the properties of two kinds of coal were correlated with NO_x formation. Combustion experiments were carried out with two kinds of coals in a pilot scale bubbling fluidized bed boiler. The results of the experiments confirm most of the findings reported in the literature, with the exception of the effect of bed temperature. The following general conclusions can be drawn:

(1.) Although GYC has a lower nitrogen content, the NO$_x$ production is higher. This is explained by the higher reactivity of GYC and by the higher volatile content, which is about 10 % higher than the value for CBC.

(2.) The two fuels have a similar correlation with the oxygen concentration in the bed, and with the primary/secondary air ratio. An increase in both parameters increases the production of NO$_x$.

(3.) The production of NO$_x$ from CBC combustion depends strongly on the fluidization velocity. The NO$_x$ emissions also increase with increasing fluidization velocity. For GYC, the dependence is less significant.

(4.) For both fuels, no clear dependence was found between production of NO$_x$ and bed temperature.

Acknowledgements

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