



## Experimental investigation of NO<sub>x</sub> emissions in oxycoal combustion

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### ABSTRACT

This work presents the results of an experimental investigation on NO<sub>x</sub> emissions from coal combustion in a pilot scale test facility. Three oxidiser atmospheres have been compared, namely air, CO<sub>2</sub>/O<sub>2</sub>, and O<sub>2</sub> enriched recirculated flue gas. NO<sub>x</sub> emissions from two different combustion modes have been studied, swirl flame and flameless combustion. The influence of the burner oxygen ratio and the oxidiser O<sub>2</sub> concentration on NO<sub>x</sub> formation and reduction have been analysed. With increasing burner oxygen ratio, an increase of NO<sub>x</sub> emissions has been obtained for air and CO<sub>2</sub>/O<sub>2</sub> in both, swirl flame and flameless combustion. In case of the swirl flame, flue gas recirculation leads to a reduction of NO<sub>x</sub> emissions up to 50%, whereas in case of flameless combustion this reduction is around 40% compared to CO<sub>2</sub>/O<sub>2</sub>. No significant impact of the oxidiser O<sub>2</sub> concentration in the CO<sub>2</sub>/O<sub>2</sub> mixture on NO<sub>x</sub> emissions is observed in the range between 18 and 27 vol.% in swirl flames. An analysis of NO<sub>x</sub> formation and reduction mechanisms showed, that the observed reduction of NO<sub>x</sub> emissions by flue gas recirculation cannot be attributed to the reduction of recirculated NO<sub>x</sub> alone, but also to a reduced conversion of fuel-N to NO.

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### 1. Introduction

Combustion of fossil fuels results in the formation of oxides of nitrogen (NO<sub>x</sub>), either by fixation of atmospheric nitrogen (via the thermal or prompt NO mechanisms for NO formation), or by conversion of nitrogen containing structures in the fuel (the fuel NO mechanism). Due to the relatively high content of nitrogen in coal and its extensive use as fuel, coal combustion is a major source of anthropogenic NO<sub>x</sub> emissions, and mechanisms for formation of NO<sub>x</sub> from combustion of coal have been extensively studied. Combustion of coal under controlled conditions has demonstrated that NO production via the fuel NO mechanism dominates under the regime of temperatures and residence times found in full-scale pulverised fuel fired furnaces [1,2]. There are two opposing factors that influence fuel NO<sub>x</sub> emissions during coal combustion: (i) the oxidation of fuel-N by oxygen and other oxidising agents, and (ii) the reduction of already produced NO<sub>x</sub> by reducing agents, such as hydrocarbons from pyrolysis of the volatile matter (homogeneous reactions) and resident char (heterogeneous reactions).

Numerous technologies are available nowadays to control NO<sub>x</sub> formation and reduction inside pulverised fuel fired furnaces. These methods, known as primary measures, differ significantly in cost, effectiveness, complexity, and extent of modifications required to achieve a reduction of NO<sub>x</sub> emissions. A combination of those is commonly used to comply with regulatory emission limits.

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With oxyfuel combustion, that is burning of fuel in a mixture of oxygen and recycled flue gases, see also [3–5], NO<sub>x</sub> emissions from pulverised fuel boilers are significantly reduced. Compared to air-fired units, NO<sub>x</sub> emissions generated per energy released during oxyfuel combustion are around 60–70% lower depending on the burner design, coal type and operating conditions [5–10]. Moreover, according to Normann et al. [9], conventional primary control measures should be sufficient to reduce NO<sub>x</sub> emissions in oxyfuel firing to meet equivalent regulatory limits. This substantial NO<sub>x</sub> decrease is presumed to be the result of the two following mechanisms [11,12]. Firstly, thermal NO formation is reduced due to the absence of atmospheric N<sub>2</sub>. Secondly, recycled NO is reduced to form molecular nitrogen after being supplied through the flame: (i) by CH fragments from the pyrolysis of volatile matter; (ii) by interaction between recycled NO and released fuel N, mainly HCN and NH<sub>3</sub>, and (iii) catalytically on the char surface. According to Mendiara and Glarborg [13], an increase of the CO<sub>2</sub> concentration affects the ratio of the concentrations of radicals (O/H/OH), thus leading to an increased probability of formation of N<sub>2</sub> from NH<sub>3</sub> instead of NO.

Park et al. [14] studied the fuel-N conversion during heterogeneous reaction of bituminous coal char with O<sub>2</sub>, CO<sub>2</sub> and H<sub>2</sub>O over a broad range of temperatures, pressures and reactant gas concentrations. The results showed that char-N is converted entirely into N<sub>2</sub> when char reacts with CO<sub>2</sub>; into N<sub>2</sub> and NO when char reacts with O<sub>2</sub>; and into HCN, NH<sub>3</sub> and N<sub>2</sub> when char reacts with H<sub>2</sub>O. Previous simulations of a combustion system with high in-furnace recirculation rates [15] showed that the recirculation causes in-

creased char oxidation by CO<sub>2</sub> and H<sub>2</sub>O. Together these two studies show that the amount of char-N released as N<sub>2</sub> is rather independent of the recycling ratio as long as the flue gas is not dried, since the increased N<sub>2</sub> release due to oxidation by CO<sub>2</sub> is compensated by the increased release of intermediate species by oxidation with H<sub>2</sub>O. Supposedly, char-N to NO conversion can thus be influenced by drying of the recycled flue gas and thereby adjusting its vapour content. Moreover, Cao et al. [16] showed that a high concentration of nitrogen intermediates in the combustion zone, which can be achieved by wet recycling, is favourable for NO reduction.

Next to conventional reduction techniques, Liu and Okazaki [17] as well as Normann et al. [9] proposed an increase of the combustion temperature as a means to facilitate NO<sub>x</sub> reduction via the thermal path.

The conversion of fuel-N to NO<sub>x</sub> increases with the O<sub>2</sub> concentration in the oxidiser [18–20]. However, Shaddix and Molina [19] showed that this effect is less pronounced or even reversed at O<sub>2</sub> concentrations below 24 vol.%. According to Hu et al. [12] and Liu and Okazaki [17], the NO reduction efficiency increases with equivalence ratio and recycling ratio. As the recycling ratio increases, the heterogeneous reaction between NO and char is promoted in the early stages of coal combustion by high CO concentrations, which are obtained as a result of the low O<sub>2</sub> content in the inlet gas. However, the resulting decrease of fuel concentration has an inverse effect on the reduction of NO<sub>x</sub> because of the decrease in hydrocarbon fragment concentrations. Hence, there is a contrary effect of O<sub>2</sub> content on the emission of fuel NO and the reduction of recycled NO<sub>x</sub>.

This paper focuses on NO<sub>x</sub> emissions as function of the burner oxygen ratio defined according to the following equation:

$$\text{oxygen ratio} = \lambda = \frac{m_{\text{O}_2}/m_{\text{fuel}}}{(m_{\text{O}_2}/m_{\text{fuel}})_{\text{stoichiometric}}}$$

and furthermore on NO<sub>x</sub> emissions as function of the O<sub>2</sub> content in the oxidiser. The oxygen concentration has been varied between 18 and 27 vol.% in the oxidiser as the required oxygen concentration depends on the process topology of the oxyfuel power plant [3,21]. Three different combustion atmospheres were considered, namely, air, CO<sub>2</sub>/O<sub>2</sub>, and O<sub>2</sub> enriched recycled flue gas. Furthermore, experiments have been carried out with a swirl burner which has been optimised for oxyfuel combustion as well as with a burner for flameless combustion. The motivation for this work is to investigate in more detail the processes of NO<sub>x</sub> formation and reduction in oxy-combustion of pulverised coal at oxygen concentrations similar to those in air. Moreover, the differences in NO<sub>x</sub> emissions from combustion in air, CO<sub>2</sub>/O<sub>2</sub> and a flue gas recycle mode are evaluated.

## 2. Experimental setup

### 2.1. Test facility

Experiments were conducted in a vertical, cylindrical, top fired furnace with an inner diameter of 400 mm and a total length of 4200 mm, Fig. 1. In the centre of the furnace, 2100 mm from the top, four ports give access to the combustion chamber for optical and probe measurements. The burner is axially traversable allowing for measurements at different distances from the burner through a single measurement plane. Alternatively, the measurement ports can also be used for the injection of staging air when the burner is traversed to higher positions. Moreover, the test facility is equipped with an electric heating system with which the wall temperature can be regulated. A schematic of the furnace with peripheral equipment is given in Fig. 2.

The test facility can be operated in three different modes: air, dry oxyfuel and wet oxyfuel. Air and dry oxyfuel refer to

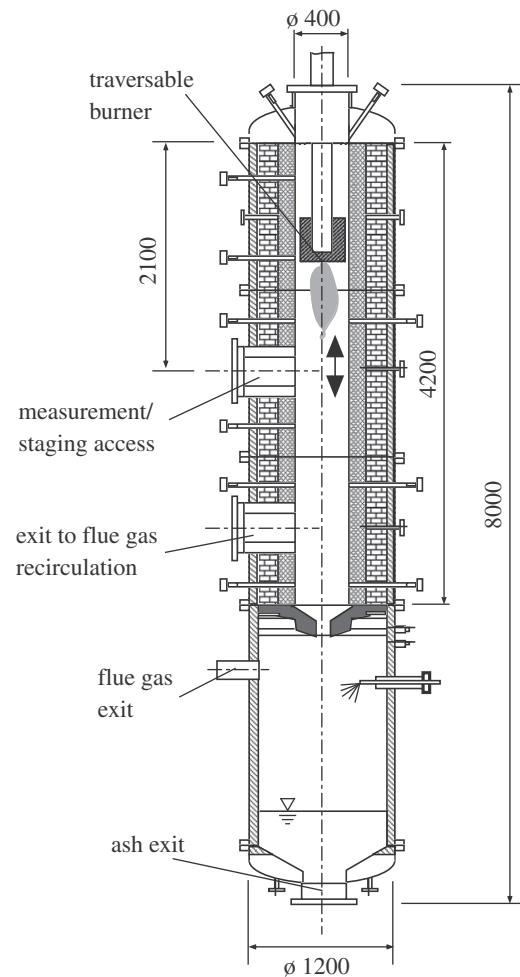


Fig. 1. Furnace geometry, all dimensions in [mm].

once-through-modes with air (point 12 in Fig. 2) or respectively a mixture of CO<sub>2</sub> and O<sub>2</sub> as oxidiser (point 7). Wet oxyfuel refers to a flue gas recycling (FGR) mode, where a part of the flue gas is recycled and enriched with oxygen up to a concentration of 21 vol.%. First of all, fly ash is removed in the recirculation (point 2), then the flue gas is enriched with oxygen (point 3), cooled to a manageable fan inlet temperature by a flue gas cooler (point 4) and fed to a reservoir (point 6) by a flue gas fan (point 5). The oxygen concentration in the oxidiser stream in dry oxyfuel mode has been varied between 18 and 27 vol.% and in wet oxyfuel between 18 and 21 vol.%.

In air mode, coal is either carried by air (with a reduced oxygen content of 19 vol.% due to inertisation of the coal feeding system – point 8 in Fig. 2) or by nitrogen. In dry oxyfuel mode, the oxygen concentration in the coal carrier is limited to a maximum of 21 vol.% in a mixture of O<sub>2</sub> and CO<sub>2</sub>. As an alternative, pure CO<sub>2</sub> can also be used as coal carrier. In wet oxyfuel mode, coal is carried by a mixture of CO<sub>2</sub> and O<sub>2</sub> with 19 vol.% O<sub>2</sub>.

### 2.2. Burners

Two different burners have been used in this investigation: one swirl burner and one burner for flameless combustion. The burner geometries and dimensions are shown in Fig. 3.

The swirl burner is a scaled up version of the burner presented in detail by Toporov et al. [22]. It has an annular orifice through which the coal and carrier stream are injected. The part of the

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