NO formation and emission under oxy-fuel combustion conditions

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Abstract
This study concentrated on the formation and emission of NO in atmospheres characteristics of oxy-fuel combustion. The investigation was conducted in a laboratory furnace fired with methane, doped with NH3 to simulate the fuel bound nitrogen present in many solid fuels, particularly in coal. The results reported include flue-gas composition data for various furnace operating conditions that, all together, allowed to evaluate the influence of the velocity and composition of the oxidiser and, also, of the initial concentration of NH3 in the fuel on NOx emissions. The results showed that (i) for a given NH3 concentration in the fuel, the oxidizer velocity has a modest effect on the NOx emissions, regardless of the oxidizer composition, presumably because the relative small variations in the oxidizer velocities are insufficient to alter significantly the main characteristics of the flow, namely the residence time; (ii) the oxidizer composition has a marginal influence on NO formation and emission in the present combustor, but it does affect the NH3 to NO conversion rates, the conversion rates increase as the O2 concentration in the oxidizer increases; (iii) increasing the NH3 concentration in the fuel yields higher NOx emissions simply because more NH3 is available to be converted to NO; higher NH3 concentrations in the fuel originate however lower conversion ratios.

Keywords: Experimental, oxy-fuel combustion, oxidizer composition, NO formation; NOx emissions

1. Introduction
For coal to continue to drive power generation and economic expansion across the world in the most sustainable manner, technology must continue to be developed in order to reduce coal power plants emissions to near-zero. Among the carbon capture and storage technologies, oxy-fuel combustion is seen as one of the most promising, with a number of recent studies showing its potential to reduce CO2, NOx and SO2 emissions. Oxy-fuel combustion consists of combustion in mixtures of O2/CO2 rather than air, being in practical applications the O2 obtained from the atmospheric air with the aid of a separation unit and the CO2 from the recycling of part of the flue gas.

A large number of studies have been carried out in the last years in order to apply this technology to power generation plants, from where more than one third of the global CO2 emissions come from. More than 10 years ago, Takano et al. [1] evaluated the effect of O2/CO2 atmospheres on ignition, flame speed and NOx and SO2 emissions in an industrial furnace fired with bituminous Japanese coal. The authors observed conversion ratios of fuel bound nitrogen to NO above 30% for air combustion without fuel staging, while very low conversion ratios were obtained under oxy-fuel combustion conditions with flue gas recirculation. They attributed these results to the reburning mechanism that occurs during flue gas recirculation. Thambimuthu and Croiset [2] examined the emissions of NOx and SO2 under air and oxy-fuel combustion conditions using also coal as fuel in a 0.21 MW furnace. The results revealed that combustion in air presented the highest NOx emission rates due to the NO thermal formation, while combustion with recycled flue gas yielded lower NOx emission rates than for once-through combustion in O2/CO2 mixtures. This effect was explained by the reburning mechanism activity, which led to 40%-50% reduction in NOx emissions, as compared to the non flue gas recirculation cases. Normann et al. [3] carried out an experimental and numerical study in a lignite fired 100 kW furnace centered on the effect of temperature, residence time, stoichiometry and dry flue gas recirculation rate on NO formation. The results confirmed the significant influence of the reburning mechanism on NOx reduction, as observed earlier by Ando and Okazaki [4], with 50% to 80% of the NO to N2 reduction occurring in the combustion zone. Takano et al. [5] obtained earlier on similar NOx reductions, which were explained by the high HCN and NH3 concentrations found in the near burner region during operation under oxy-fuel combustion, in contrast with combustion in air. Normann et al. [3] concluded also that the influence of the stoichiometry in the NO formation was marginal, in accordance with the findings of Takano et al. [1], who concluded that NOx emissions could be reduced by about 50% and 70% under oxy-fuel combustion conditions, as compared to the air-firing case.

Andersson et al. [6] studied numerically the NOx reduction potential under oxy-fuel combustion conditions for high-temperatures. Methane was used as fuel, with small amounts of HCN and NH3 added to simulate the fuel bound nitrogen. In addition to CO2, H2O and NO, the oxidizer included also N2 to simulate air leakages into the system. The Zeldovich mechanism was isolated from other mechanisms (e.g., reburning) and results revealed that the Zeldovich mechanism remains inactive for temperatures below 1400 °C, while the reburning mechanism is the dominant phenomenon. However, for higher temperatures, NO reduction is dominated by the inverse Zeldovich mechanism. Results also
showed that for sufficiently high N\textsubscript{2} concentrations, the reverse thermal mechanism stops acting. Instead, NO formation takes place as in air combustion.

Recently, Zhang et al. [7] examined the NO\textsubscript{x} emissions under oxy-fuel combustion conditions using two bituminous coals in O\textsubscript{2}/N\textsubscript{2}, O\textsubscript{2}/Ar, O\textsubscript{2}/CO\textsubscript{2}, and O\textsubscript{2}/recirculated flue gas atmospheres, with O\textsubscript{2} initial content in the oxidizer varying from 8% up to 40%. The authors observed that increasing the O\textsubscript{2} content in the oxidizer increased the conversion ratios of the fuel bound nitrogen to NO for all tested atmospheres, which is consistent with the results of Molina and Shaddix [8]. Zhang et al. [7] reported that the lowest conversion ratios occur for the O\textsubscript{2}/recirculated flue gas atmospheres. This was attributed to the absence of atmospheric nitrogen, high CO\textsubscript{2} concentration and NO reduction through flue gas recirculation. The results also showed that key factor for the lower conversion ratios in O\textsubscript{2}/recirculated flue gas atmospheres is the NO reduction through flue gas recirculation, being more important for higher flue gas recirculation rates. However, the importance of this factor decreases as the O\textsubscript{2} concentration in the oxidizer increases.

Christ et al. [9] performed similar tests in a 40-100 kW furnace. The authors used swirl and non-swirl burners and operated the combustor also under oxy-fuel flameless combustion conditions. Wet and dry flue gas recirculation have also been used. The results showed that increasing the O\textsubscript{2} in the oxidizer led to higher NO\textsubscript{x} emissions for all oxidizers. The results also showed a 50% NO\textsubscript{x} emission reduction by using flue gas recirculation in the swirl burner and up to 40% under flameless combustion operation, as compared to the non flue gas recirculation case. It was also concluded that wet flue gas recirculation yields significantly lower emission levels, as compared with dry flue gas recirculation for the oxidizers studied. The authors sustain that steam is responsible for inhibiting in some way intermediate species on NO formation, and thus it is better to operate with wet flue gas recirculation in oxy-fuel combustion systems as far as the NO\textsubscript{x} emissions are concerned.

Kobayashi et al. [10] concluded that recycled NO to N\textsubscript{2} reduction efficiency increases with the flue gas recirculation rate. In general, most authors point out the reburning mechanism that takes place when recycling flue gas through burner as the responsible for the small oxy-fuel NO\textsubscript{x} emission levels, as compared to air-firing. However, it is important to analyze not only NO to N\textsubscript{2} reduction through reburning, but also the influence of the CO\textsubscript{2} rich atmosphere on NO formation from the fuel bound nitrogen. Glarborg and Mendiara [11] studied experimentally and numerically the ammonia chemistry under oxy-fuel combustion conditions. The authors used methane as fuel, studying fuel lean and rich mixtures in the temperature range of 973 K and 1773 K. The results showed that increasing the CO\textsubscript{2} content in the oxidizer affects the O/H/OH radicals concentrations leading to an extended probability of N\textsubscript{2} formation from NH\textsubscript{3}, instead of NO formation.

Finally, it is important to mention an on-going project from Babcock & Wilcox and Air Liquide [12] in a 30 MW pilot-scale facility. The results confirm a decreasing in NO\textsubscript{x} and SO\textsubscript{2} emissions, as compared to air-fired cases for all tested fuels (several coal types), reaching NO\textsubscript{x} reductions up to 70% for bituminous coal.

Despite the work mentioned above, there is a lack of detailed experiments that could provide a deep insight into the oxy-fuel combustion phenomena, particularly in regard to the oxidizer composition impact on NO formation. Such detailed experiments are only feasible on the laboratory scale, like in this work. The present study focuses on NO formation and emission under oxy-fuel combustion conditions. Specifically, the main aim of the present study is to provide a set of NO\textsubscript{x} emission data for distinct oxidizer injection velocities, distinct oxidizer compositions and distinct initial NH\textsubscript{3} concentrations in the fuel, which simulates the fuel bound nitrogen in fuels such as coal.

2. Test Facility and Experimental Methods

Figure 1 shows a schematic of the combustor used in this study. The combustion chamber is a quartz-glass cylinder with an inner diameter of 100 mm and a length of 340 mm. During the tests, the quartz cylinder was insulated with a 30-mm-thick ceramic fiber blanket. The burner and the exhaust port are mounted at the top end of the combustion chamber. The burner consists of a central orifice of 4 mm inner diameter, through which the fuel is supplied, surrounded by an annular orifice with a variable inner diameter (14, 14.5, 15 and 15.5 mm in this work) and 18.5 mm outer diameter for the oxidizer supply. The exhaust consists of an annular orifice, concentric with the burner, with 75 mm inner diameter and 90 mm outer diameter. A stainless steel plate is used to close the bottom end of the combustion chamber. This plate contains a moving hole with an inner diameter of 11 mm that allows for the introduction of probes. The oxidizer is preheated by an electrical heating system that allows inlet temperatures up to 700 °C, which are monitored by a type K thermocouple installed at the entrance of the burner.

Figure 2 shows a schematic of the fuel and oxidizer feeding systems. Methane and small amounts of NH\textsubscript{3}, from gas cylinders, are fed to the combustor as fuel. As for the oxidizer, air, from a
compressor, or mixtures of O₂, CO₂, from gas cylinders, and H₂O are supplied to the combustor as shown in Figure 2. The H₂O was supplied to the oxidizer stream either from a steam generator or by saturating the CO₂ stream through a bubbling water system at room temperature. All flow rates were measured with calibrated rotameters.

Flue-gas composition data were obtained using a stainless steel water-cooled probe. The probe was composed of a central 1.3 mm inner diameter tube through which quenched samples were evacuated. This central tube was surrounded by two concentric tubes for probe cooling. The gas sample was drawn through the probe and part of the system by an oil-free diaphragm pump. A condenser removed the main particulate burden and condensate. A filter and a drier removed any residual particles and moisture so that a constant supply of clean dry combustion gases was delivered to the analyzers through a manifold to give species concentration on a dry basis. The analytical instrumentation included a magnetic pressure analyzer for O₂ measurements, a non dispersive infrared gas analyzer for CO₂ and CO measurements, a flame ionization detector for hydrocarbons (HC) measurements and a chemiluminescent analyzer for NOₓ measurements. At the combustor exit, probe effects were negligible and errors arose mainly from quenching of chemical reactions, which was found to be adequate. Repeatability of the flue-gas data was, on average, within 10%.

3. Test Conditions

Table 1 presents the experimental conditions used in this investigation. As can be seen, five oxidizer compositions were tested, including air, with each oxidizer being injected through four different oxidizer diameters to evaluate the effect of the oxidizer velocity on NO formation and emission. Additionally, for all conditions, four small flow rates of NH₃ were injected together with fuel in order to simulate the nitrogen present in some fuel, particularly in coal. All together, the experimental conditions allowed evaluating the oxidizer injection velocity and composition as well as the initial NH₃ content in the fuel on NO formation and emission. As seen in Table 1, the O₂ mass flow was kept constant throughout the tests, while the CO₂ mass flow rate was varied in order to establish O₂/CO₂ oxidizers with distinct compositions. The table also reveals that the adiabatic flame temperature, and thus most likely the temperature inside the combustion chamber, increases as the O₂ concentration in the oxidizer increases.

4. Results and Discussion

Aerodynamic Characteristics of the Present Flames

In order to better interpret the present results, it is helpful to understand the flow aerodynamics inside the combustion chamber. Figure 3 shows the flow inside the combustion chamber, as predicted using the FLUENT by Graça [13], for test 1 in Table 1. The figure shows the stream lines on the left side and the velocity vectors on the right side. The flow is characterized by a large recirculation zone, around the central jet, that extends from the near burner region up to 2/3 of the combustor length. The combustor configuration, with the burner and the exhaust port mounted at the same end of the combustion chamber (Figure 1), causes a flow deceleration which promotes the stabilization process and enhances the mixing between the fresh reactants and the flue gas, mainly due to the high levels of turbulence. Note that the central jet penetration into the combustion chamber, the mixing intensity and the residence times decrease as the oxidizer velocity decreases.

Effect of the oxidizer velocity

Figure 4 shows the NOₓ emissions, expressed in ng/J, as a function of the oxidizer velocity for five oxidizer compositions and four NH₃ concentrations. It is seen that, for a given NH₃ concentration in the fuel, the oxidizer velocity has a modest effect on the NOₓ emissions, regardless of the oxidizer composition. This is presumably because the relative small variations in the oxidizer velocities are insufficient to alter significantly the main characteristics of the flow, namely the residence time. Despite the relatively small range of oxidizer velocities studied, this result is relevant for industrial applications, where changes in flue gas recirculation rates can alter the oxidizer velocity, which, according to the present results, yields insignificant changes in the NOₓ emissions.

Effect of the oxidizer composition

Returning to Figure 4, it is seen that, for a given NH₃ concentration in the fuel, the NOₓ emission levels are, with very few exceptions, approximately constant regardless of the oxidizer composition. Due to the absence of N₂, the fuel-NO mechanism is responsible for all NOₓ emissions under oxy-fuel combustion conditions. In air combustion, besides the fuel-NO mechanism, the thermal NO mechanism may play an important role. However, Figure 4 presents levels of NOₓ emissions similar for oxy-fuel combustion with an oxidizer with 20.9% O₂ and 79.1% CO₂ and combustion in air, which
suggests that the NO-thermal mechanism in the present air flames plays a negligible role. Despite the fuel-NO mechanism being very dependent on local O\textsubscript{2} concentration, NO\textsubscript{x} emissions, for a given NH\textsubscript{3} concentration in the fuel, are very similar for all oxidizers. This result can be partially explained by the O\textsubscript{2} mass flow rate being kept constant during all tests, see Table 1.

Figure 4 suggests therefore that the oxidizer composition has a marginal influence on NO formation and emission in the present combustor. Does the oxidizer composition affects however the NH\textsubscript{3} to NO conversion rate? In order to clarify this question, Figure 5 shows the NH\textsubscript{3}/NO ratio as a function of the O\textsubscript{2} in oxidizer for four NH\textsubscript{3} concentrations. It is seen that oxidizer composition does affect the NH\textsubscript{3} to NO conversion rates: the conversion rates increase as the O\textsubscript{2} concentration in the oxidizer increases.

Effect of the concentration of NH\textsubscript{3} in the fuel

Figure 4 also reveals that increasing the NH\textsubscript{3} concentration in the fuel yields higher NO\textsubscript{x} emissions simply because more NH\textsubscript{3} is available to be converted to NO. However, it is important to notice in Figure 5, that higher NH\textsubscript{3} concentrations in the fuel originate lower conversion ratios. This is explained by the increasing relevance of the reburning mechanism, responsible for the conversion of part of the NO to N\textsubscript{2}.

5. Conclusions

The main conclusions of the present study are as follows.

- For a given NH\textsubscript{3} concentration in the fuel, the oxidizer velocity has a modest effect on the NO\textsubscript{x} emissions, regardless of the oxidizer composition. This is presumably because the relative small variations in the oxidizer velocities are insufficient to alter significantly the main characteristics of the flow, namely the residence time.
- The oxidizer composition has a marginal influence on NO formation and emission in the present combustor, but it does affect the NH\textsubscript{3} to NO conversion rates, the conversion rates increase as the O\textsubscript{2} concentration in the oxidizer increases.
- Increasing the NH\textsubscript{3} concentration in the fuel yields higher NO\textsubscript{x} emissions simply because more NH\textsubscript{3} is available to be converted to NO; higher NH\textsubscript{3} concentrations in the fuel originate however lower conversion ratios.

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References

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Table 1. Experimental conditions. For all conditions: thermal output = 6 kW; $V_{CH_4} = 14.4$ m/s; $T_{oxidizer} = 500$ °C; $\lambda_{O_2} = 1.1$. 
Figure 1. Schematic of the combustor.

Figure 2. Schematic of the fuel and oxidizer feeding systems.
Figure 3. Flow inside the combustion chamber as predicted by Graça [13] using the FLUENT.
Figure 4. NO\(_x\) emissions as a function of the oxidizer velocity for five oxidizer compositions and four NH\(_3\) concentrations.
Figure 5. NH₃/NO ratio as a function of the O₂ in oxidizer for four NH₃ concentrations.