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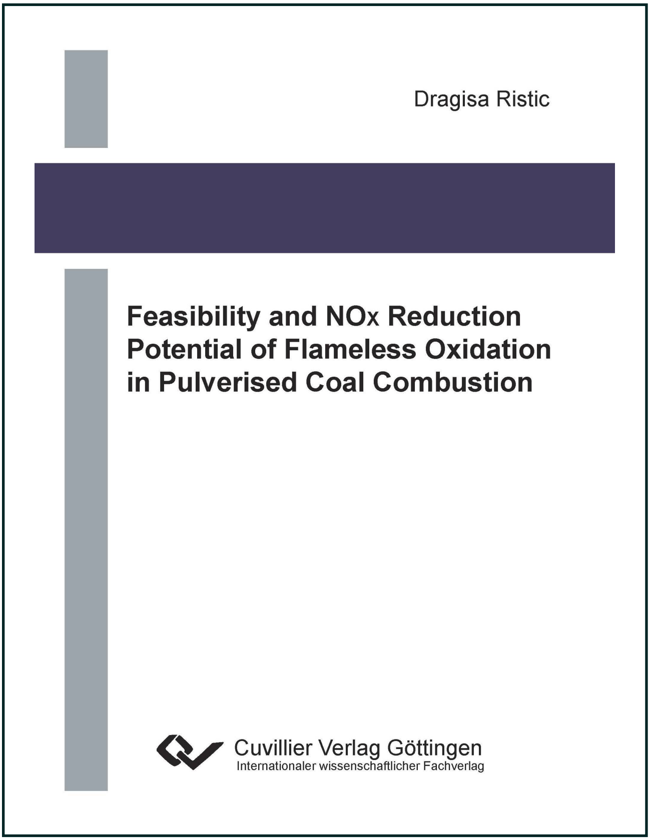
Feasibility and NOx Reduction Potential of Flameless Oxidation in Pulverised Coal Combustion



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1 Introduction

1.1 Coal combustion in power generation in response to environmental issues

Combustion of coal in steam generating boilers is the prevailing process of fossil energy utilisation in electric power generation, supplying over one third of the electricity worldwide. Due to its low-cost and broad availability, it is viewed as competitive energy source well into the twenty-first century. However, coal-fired power plants have significant environmental concerns. They are source of numerous harmful pollutants responsible for accelerated rate of global warming, acid rain, etc. Hence, the prudent public policy is to aim at the development and early application of clean-coal combustion technologies in a high efficient power cycle. According to Beér (2000), the development of combustion technologies in the electric power generation has been classified into three time periods:

- the pre-environmental era (up to 1970) in which the objectives were complete combustion with a minimum of excess air and capability of scale-up to increased boiler unit performances,
- the environmental era (after 1970) in which reduction of combustion generated pollution was receiving increased international attention, and
- the present and near future in which a combination of clean-coal combustion and high thermodynamic efficiency is considered to be necessary to satisfy the next levels of pollutant compliance and demand for CO_2 emission mitigation.

From the 1970s onwards, the applied combustion researches have taken a turn from high output and high intensity combustion towards combustion process modification and pollutant emissions control technologies. Hence, during the last decades there has been an additional task of developing highly efficient combustion processes and pollutant reduction systems to be able to meet demands for reduced pollutant emissions in coal-fired power plants. Accordingly, the present coal-fired power plants have to satisfy following demands: low NO_x and SO_x emissions, cleaning up flue gas from fly ash, high total efficiency well above 40 % and in the near future control of CO_2 emission.

Nitrogen oxides remain one major pollutant from pulverised coal-fired power plants and have increased international attention because of their wide ranging effects on the environment, including their contribution to acid rain, photochemical smog, production of tropospheric ozone and, in the case of N_2O, depletion of stratospheric ozone. More stringent pollutant emission requirements create the need for improvements of NO_x emissions from coal-fired power plants. Hence, the control of primary (in-furnace) NO_x emissions has become an important aspect of the pulverised coal-fired steam generating boiler designs. Formation of NO_x is dependent not only on coal characteristics, but also on boiler and burner designs and operating conditions. Therefore, technical solutions for the reduction of NO_x ranging from air staging techniques and low-NO_x pulverised coal burners to high-cost Selective Catalytic Reduction (SCR) systems are required. The development of the low-NO_x pulverised coal burners and air staging techniques reduced considerably primary NO_x emissions in coal-fired boilers. Nowadays, supercritical or ultra-supercritical boilers equipped with commercially available NO_x reduction technologies are state of the art in pulverised coal-fired power plants. Such boilers operate in supercritical pressure conditions with considerably reduced primary NO_x emissions. However, the NO_x removal from the exhaust gas, e.g. by SCR systems, is still required in order to meet prescribed emission standards.

Concerning SO_x capture, the state of the art in pulverised coal-fired power plants is the integration of wet flue gas desulphurisation systems (scrubbers), where SO_x emissions are reduced to meet required levels. In the 1980s, development of the fluidised bed combustion provided the opportunity to effectively capture SO_x without external emission controls (e.g. scrubbers). Moreover, commercial fluidised bed combustion boilers operate at competitive efficiencies and have NO_x and SO_x emissions below currently prescribed standards. The fluidised bed combustion is, however, primarily intended for utilisation of low-quality coals in smaller capacity boilers. However, in the last decade, the development of circulated fluidised bed combustion facilitated the possibility of applying the fluidised bed technology to large single boiler units (e.g. 460 MW_{el} world's largest circulated fluidised bed boiler in Lagisza, Poland).

The CO_2 management from coal-fired power plants is becoming increasingly important in the last decade. Namely, the CO_2 emission control is gaining increasing acceptance as a result of the international global warming debate. Carbon capture and storage is the critical enabling technology to help reduce CO_2 emission significantly. Carbon capture and storage technologies might bring their opportunities for enhancing the environmental performance of coal-fired power plants in a carbon-constrained world.

Concerning fine organic and inorganic particulates, electro-static precipitators (ESP) continue to be excellent devices for cleaning up flue gases. The electro-static precipitator is highly efficient filtration device that minimally impedes the flow of gases through the device, and can easily remove fine particulate matter such as dust and smoke from the flue gas. Therefore, an electro-static precipitator applies energy only to the particulate matter being collected and is very efficient in its consumption of energy.

Flue gas treatment technologies are available for NO_x, SO_x and particles enabling an emission reduction to the mandated values, while also allowing coal to meet the world's pressing energy needs. However, these technologies require additional energy, increased operational costs of a power plant and by-products like wastewater, gypsum and used catalysts are generated. Therefore, modification of the combustion process can help in improving the combustion efficiency and reduction of the in-furnace pollutant emissions. One technology that is capable of reducing primary NO_x emissions and increasing efficiency in gaseous fuels combustion is the so-called Flameless Oxidation (FLOX^{TM}[1]). It is already widely used in the heat treatment industry with gaseous fuels, but its adaptation to pulverised coal combustion requires further investigations. Therefore, the feasibility of a Pulverised Coal Flameless Oxidation (PC-FLOX^{TM}) burner technology has to be demonstrated and potential of the PC-FLOX^{TM} burner to reduce NO_x emissions from pulverised coal combustion has to be determined.

1.2 Flameless oxidation, development and potential in pulverised coal combustion

During the 1980s, one effective way to boost thermal efficiency from gas-fired process furnaces was to highly preheat combustion air (e.g. about 1000 °C), but as a consequence NO_x emissions were also increased due to the increased production of thermal NO under high temperature. Since in conventional flames highly preheated combustion air is conducive to the both aims, i.e. reduction of NO_x emissions is in conflict with promoting the combustion efficiency. In the late 1980s and early 1990s, a flame dilution technology based on the internal flue gas recirculation has been investigated in order to suppress thermal NO formation under high temperature combustion processes, even when highly preheated combustion air is used. In Japan, at Tokyo Gas this energy efficient and environmentally friendly combustion technology has been investigated (Nakamachi et al., 1990). The concept has utilised an idea of discrete injection of fuel gas into hot combustion products (i.e. flue gas), and hence, a flame dilution has been facilitated.

In the late 1990s, Katsuki and Hasegawa (1998) investigated a combustion system with air preheat up to 1000 °C and revealed a high dilution of the reactants followed by low NO_x emissions. Due to the high inlet air temperature this technology has been named "High Temperature Air Combustion (HiTAC)". Contemporarily in Europe, the company Wärmeprozesstechnik (WS) GmbH was the major developer of this combustion technology based on the flame dilution. Hence, as a result of this research, the technology of the "Flameless Oxidation" or "FLOX^{TM}" has been developed and patented worldwide by WS GmbH, Renningen, Germany. Since the

[1] The acronym FLOX^{TM} is a registered trademark of WS Wärmeprozesstechnik GmbH, Germany.

FLOX^{TM} technology does not require highly preheated combustion air, the naming was based on the characteristics of the flame itself.

The flame dilution means that fuel and combustion air are mixed with a ballast of internally recirculated flue gases, which creates oxygen diluted conditions (i.e. decrease in oxygen partial pressure) before the self-ignition temperature of the fuel is attained. Internal recirculation of flue gases, or the so-called combustion products from inside the furnace back to the combustion zone carries out the most common dilution mechanism. Therefore, oxygen concentration in the reactants is substantially reduced with respect to the 21 vol.-% of the standard oxidising air. The dilution of the reactants leads to a volumetric combustion, where chemical reactions occur within a large volume of the furnace instead of the distinct flame fronts with high temperature peaks found in conventional combustion flames. Overall, in flameless oxidation the oxidation of fuel occurs at temperatures above the self-ignition temperature with a very limited oxygen supply. Spontaneous ignition occurs and progresses with no visible or audible signs of the flame. The chemical reaction zone is quite diffuse, and this leads to almost uniform heat release and a smooth temperature profile. Herewith, formation of thermal NO_x is effectively abated. This has given rise to a definition of FLOX^{TM} already published by its inventors (Wünning and Wünning, 1997; Wünning, 2005):

> ”The combustion at the flameless oxidation is mixture and temperature controlled, achieved by specific flow and temperature conditions, it is a stable combustion without a visible flame and with defined internal recirculation of a hot inert flue gases”.

Back in the 1970s, Hardesty and Weinberg (1974) described already a combustion technology for gases of low-heat content, where the enthalpy of the reactants is increased due to the heat recirculation. At that time, the technology has been named “excess enthalpy combustion” while today it is often called:

- Flameless Oxidation (FLOX^{TM}) (Wünning and Wünning, 1997),
- High Temperature Air Combustion (HiTAC) (Katsuki and Hasegawa, 1998), and
- Moderate and Intensive Low oxygen Dilution (MILD) combustion (Cavaliere and de Joannon, 2004).

Wünning and Wünning (1997) investigated the domain of the flameless oxidation with natural gas combustion, as a function of the recirculation ratio defined as ratio of recirculated mass flow of the combustion products (before reaction) with respect to the driving flow rate of reactants.

$$K = \frac{\dot{m}_{rec}}{\dot{m}_{air} + \dot{m}_{fuel}} \tag{1}$$

They found that three combustion modes arise from the variation of flue gas recirculation ratio and furnace temperature (Fig. 1). Stable flames (A) are possible over a whole range of furnace temperature with recirculation rates <0.3. Stable flames change into unstable turbulent flames (B) when the flue gas recirculation ratio increases. A further increasing of the ratio extinguishes the combustion reaction for temperatures below the fuel self-ignition temperature, whereas the combustion reaction enters the flameless mode (C) for high flue gas recirculation ratios and high combustion temperatures. For temperatures >850 °C, i.e. above self-ignition, the flameless oxidation can be established, corresponding to large flue gas recirculation ratio >3 that is obtained with high momentum of the injected fuel and combustion air into the furnace.

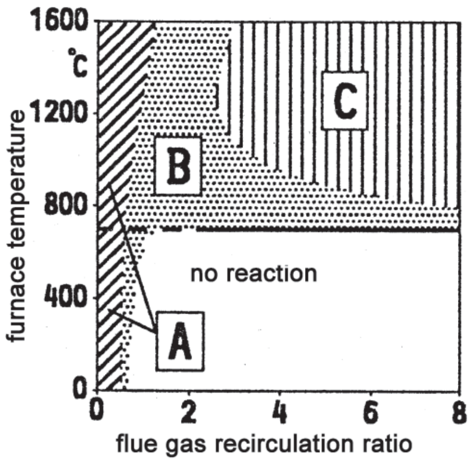

Fig. 1. **Influence of temperature and flue gas recirculation on flame stability limits (Wünning and Wünning, 1997).**

Furthermore, it is not possible to operate in flameless oxidation mode in a cold furnace. Therefore, the furnace must be heated up with flame combustion mode and then could be switched to flameless oxidation mode. A special feature of the gas-fired flameless oxidation burners is a high internal flue gas recirculation achieved in an integrated single burner design without any additional measures. Hence, due to the high recirculation ratio the air preheating is no longer a requirement for switching into the flameless oxidation mode, especially for gaseous fuels with a high-heating value.

In the late 1990s, the FLOX^{TM} technology, in particular with gas firing, has been developed for industrial use exhibiting ultra-low NO_x emissions and substantial energy savings. The technology found a number of applications mainly in steel industry and there are a number of other applications emerging. There is a large potential in ceramic, glass and chemical industries, as well as in electric power generation. Today, flameless oxidation is one of the most rapidly developing combustion technologies as a measure to reduce NO_x emissions from combustion processes. It is the state of the art for gas burners in the heat treatment industry in which, exhaust NO_x emissions are near to the detection limit.

As mentioned before, the dilution of the fuel and air mixture by the intense flue gas recirculation decreases local oxygen concentration. Therefore, in case of coal e.g. will minimise the contact of NO_x precursor compounds (such as HCN, NH_3 or char bound nitrogen) and oxygen, which could lead to the reduction of fuel NO formation. In a regenerative high temperature air burner, this ability to decrease fuel bound nitrogen (fuel-N) conversion to fuel NO has been shown for Kerosene and liquefied petroleum gas combustion (Shimo, 2000). Hence, knowing that fuel NO is dominant in pulverised coal combustion, the application of FLOX^{TM} technology to the combustion of pulverised coals has become a subject of increased interest. For most practical cases of pulverised coal combustion the fundamental reactions involved in formation and destruction of the fuel and thermal NO are known. The quantity of the formed fuel NO is dependent on the influence of burner and furnace conditions, and the properties of coal being fired. It is also known that the dilution of the combustion zone with recirculating flue gas leads to a NO reburning, the mechanism capable of reducing fuel NO to molecular nitrogen (N_2) through reactions between NO and hydrocarbon free radicals (Miller and Bowman, 1989; Smoot et al., 1998). Furthermore, internal recirculation of the flue gas provides an environment in the combustion zone that is rich in carbon monoxide, char and hydrocarbon radicals. Such environment in the combustion zone is typical of the air-staged combustion, the technology which has been established in the middle 1970s as an effective means of reducing fuel NO formation.

Overall, based on the progress in understanding of the NO_x chemistry involved in pulverised coal combustion processes, the flameless oxidation technology in the last decade is receiving increased attention as a perspective measure to reduce primary NO_x emissions from pulverised coal-fired utility boilers. Moreover, the problem of high primary NO_x emissions could be solved by applying the flameless oxidation technology in combination with already existing NO_x reduction technologies (e.g. air staging). Hence, this could avoid an expensive installation or retrofit of a SCR system in order to meet various emission standards. The flameless oxidation technology is able to combine all the requirements of modern burner designs: environmentally friendly, stable and reliable operation, excellent combustion stability and efficiency. This might lead to a significant economic advantage and improvement of the use of coal as a clean energy source compared to already existing pulverised coal burners.

The PC-FLOX^{TM} burner technology to be demonstrated at a pilot-scale (100's of kW_{th}), evaluated as being feasible and the technology's NO_x reduction potential determined are suitable ways to strengthen the further development to the full-scale (10's of MW_{th}).

1.3 Tasks and objectives of the present work

The present work aims at the development and demonstration of a 300 kW_{th} pilot-scale PC-FLOX^{TM} burner for low-NO_x pulverised coal combustion at a 0.5 MW_{th} combustion test facility. Overall scientific and technical objectives of the pilot-scaled PC-FLOX^{TM} burner demonstration are twofold. At first, feasibility of the technology is to be evaluated, and then the potential of the burner to reduce NO_x emissions from pulverised coal combustion is to be determined.

To overcome current limitation concerning NO_x reduction of an existing pulverised coal bench-scale flame burner, a 10 kW_{th} bench-scale PC-FLOX^{TM} burner design with a novel integrated air and fuel injection scheme is developed, based on the state of the art gas FLOX^{TM} burner. The bench-scale PC-FLOX^{TM} burner is tested with different pulverised coals and under various operational conditions. In a next step, based on the bench-scale experimental experience, literature study and basic numerical modelling of the pilot-scale PC-FLOX^{TM} burner is developed. Substantial pilot-scale experimental trials are conducted in order to demonstrate feasibility and NO_x reduction potential of the technology. These studies could be the backbone for the further development of PC-FLOX^{TM} burner technology to the full-scale.

Development of the PC-FLOX^{TM} burner is a challenging task since pulverised coal follows different mechanisms of combustion, and hence, flameless oxidation concepts which work for gaseous fuels cannot be directly adapted. In the case of coal e.g. the combustion is much more complex due to the succession of distinct phases, i.e. particle drying, devolatilisation, combustion of volatiles and char burnout. In addition, these mechanisms also overlap in time, at least partly. Therefore, it was essential for the success of the pilot-scale PC-FLOX^{TM} burner experimental trials that research, development and demonstration are strongly supported by fundamental bench-scale experimental studies. Besides the pilot-scale experimental trials being a final stage of the present work, the work focuses primarily on the fundamental experimental studies at a bench-scale test facility necessary for:

- understanding pulverised coal combustion behaviour in the flameless oxidation conditions, and
- improvement of the PC-FLOX^{TM} burner design.

Experiences and know-how gained from these studies supported the PC-FLOX^{TM} burner scale-up to the pilot-scale. Both test facilities, bench- and pilot-scale, which are used are duly equipped and provide a good environment to meet the needs of the experimental research. The central tasks of the present work are:

Experimental study on pulverised coal flameless oxidation process: The bench-scale PC-FLOX^{TM} burner is tested under various operational conditions in order to optimise the operational behaviour with respect to coal burnout, pollutant emissions and NO_x

formation. Reduction of NO_x emissions from gaseous fuels combustion is a significant advantage of the flameless oxidation technology. Hence, detailed study is performed in order to identify and compare reaction mechanisms forming and reducing NO in the pulverised coal flameless oxidation and flame combustion. Detailed profile measurements of important NO intermediate compounds such as HCN and NH_3 are conducted in order to investigate the quantitative conversion of fuel-N to fuel NO. Investigation of NO formation under combustion in argon-oxygen atmosphere (Ar/O_2=79/21 vol.-%) is carried out in order to quantify the ratio of fuel to thermal NO. Objectives of this part of the present work are to investigate burnout in pulverised coal flameless oxidation and potential of the flameless oxidation technology to reduce NO formation through both the fuel and the thermal mechanisms.

Optimisation of the near burner aerodynamics, experimental study: This part of the experimental research is aimed finding best burner design configuration with respect to the combustion efficiency and low-NO_x emissions. To significantly reduce NO_x emissions ensuring at the same time high combustion efficiency a near burner aerodynamics should be optimised. Therefore, based on the gained experience in the previous experimental study several burner designs with different injection schemes (coal/primary air and secondary air) are designed and experimentally tested. The experimental study is conducted with different coal types at a bench-scale combustion test facility in air-staged (fuel-rich) and unstaged (fuel-lean) combustion conditions.

PC-FLOX™ burner scale-up: Existing scale-up criteria are found to be inadequate for burners which operate with a high flue gas internal recirculation ratio (Kumar et al., 2005). Therefore, on the basis of several existing scale-up criteria and with help of a basic numerical modelling, a scale-up methodology is evolved and suggested. The objectives of the numerical modelling are to optimise the burner geometry, to quantify recirculation ratio, to predict the flow field and the combustion behaviour of a pilot-scaled PC-FLOX™ burner. The modelling will be performed with the CFD three dimensional combustion simulation AIOLOS code. The central objective of this task is to scale-up PC-FLOX™ burner to the pilot-scale. The pilot-scaled PC-FLOX™ burner design should be able to facilitate wide spread parametric investigations in order to fulfil scientific and technical objectives of the present work.

Demonstration of the pilot-scale PC-FLOX™ burner: The pilot-scaled PC-FLOX™ burner is tested at a pilot-scale combustion test facility in order to demonstrate feasibility of the pulverised coal flameless oxidation and to determine the NO_x reduction potential of the burner. The burner is tested with lignite and bituminous coal under various operational conditions in order to optimise the operational behaviour with respect to coal particles ignition, coal burnout, pollutant formation and emissions. During these experiments, besides continuous measurement of the furnace exhaust gas composition, detailed in-flame measurements of the main gas species as well as gas temperature are conducted.

1.4 Approach and outline of the present work

First, an overview of the state of the art is given. Within the overview short history on investigation of pulverised coal flameless oxidation as well as investigations on pulverised coal combustion under high-temperature air is described. Furthermore, the state of the art on NO formation and reduction mechanisms in pulverised coal combustion is given as a closure of Chapter 2.

In Chapter 3, bench- and pilot-scale combustion test facilities that were used in experimental studies are presented. Furthermore, measurement equipment is presented with a calibration description and error discussion. Additionally, descriptions of the used coals' main characteristics together with proximate and ultimate analyses are presented.

In Chapter 4, experimental study on the pulverised coal combustion behaviour in flameless oxidation is discussed. The beginning of the chapter contains the description of the proposed bench-scale PC-FLOX^{TM} burner. Hereafter, investigations on carbon and coal burnout, NO formation and reduction, and NO_x emissions in the bench-scale experimental study are presented. For all the flameless oxidation experiments reported here, the results are compared with the reference flame combustion tests and discussed.

Chapter 5 is devoted to the bench-scale experimental investigations on the optimisation of the near burner aerodynamics, especially regarding the mixing between the coal and secondary air. Effects of the burner configuration, i.e. injection scheme of the primary and secondary air, on NO reduction potential and combustion efficiency are estimated and discussed.

In Chapter 6, an overview of different burner scaling criteria is given together with summary of the pilot-scale PC-FLOX^{TM} burner geometrical dimensions derived from these criteria. As the numerical modelling supported the scale-up process, the validation of used combustion model is shown. Further, applications of the validated model to the differently scaled pilot-scale PC-FLOX^{TM} burners are described and discussed. As a closure of the chapter proposed pilot-scaled PC-FLOX^{TM} burner design is presented.

Chapter 7 contains experimental trials conducted with the developed pilot-scale PC-FLOX^{TM} burner firing lignite and bituminous coal. The results obtained during continuous exhaust gas measurements, ash sampling and detailed in-flame measurements are presented. Feasibility of the PC-FLOX^{TM} technology, its combustion efficiency and potential to reduce NO_x emissions are evaluated and discussed. Furthermore, plots of the detailed in-flame measurements of the main gas species and gas temperature in the combustion zone are presented within this chapter.

Finally, in the summary and conclusion, the major results are discussed with respect to the potential of PC-FLOX^{TM} burner technology to reduce NO_x emissions. Further perspectives for improvement of PC-FLOX^{TM} technology and burner scale-up to the full-scale are outlined.