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OVERVIEW OF THE GAZ DE FRANCE R&D ACTIVITIES ON FLAMELESS OXIDATION APPLIED TO HIGH TEMPERATURE PROCESSES

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ABSTRACT

This paper presents the work that has been done at Gaz de France's Research and Development Division for the past thirteen years on flameless combustion. This work started with the characterisation of flameless regenerative burners in our labs in collaboration with burner manufacturers. The main conclusion of the tests is that these burners are very low NOx (less than 300 $mg/m^3(n)$ at $3\%O_2$), whatever the operating conditions, while having a very high thermal efficiency (more than 70%). Gaz de France initiated a project with joint funding from Arcelor Research, Stein-Heurtey (furnaces manufacturer) and the French Environment Agency (Ademe), to evaluate the capabilities of the HiTAC (High Temperature Air Combustion) technology. This project, called InterNOx, ended in November 2003 was composed of three phases: basic understanding of this combustion mode, through semi-industrial test rigs, development of numerical simulation tools to reproduce the furnace (CFD, global tools) and demonstration operation at industrial scale. The tools developed during this project enable us to help the installation of such burners on industrial furnaces. Additional work is in progress with the CORIA laboratory (Rouen) to study flameless combustion at laboratory scale through the building of a laboratory facility allowing the study of confinement, very precise laser diagnostics and combustion of by-product gases. Moreover, an original simulation approach was carried on at Gaz de France to model flameless oxidation with a network of ideal reactors, coupled with detailed chemical kinetics. Over studies are in progress to evaluate the opportunity of installing such technologies in other industrial sectors (ceramics, petrochemicals, glass melting, gas turbines, etc.).

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1. INTRODUCTION

The concentrations of NOx from first regenerative burners were known to be such that they significantly exceeded the 'achievable release levels'. For more than fifteen years now, important studies have been realized in Japan, Germany, and USA to develop new types of burners operating with high temperature combustion air (over 1000℃) while not only reducing NO_x emissions, but also increasing furnaces temperature uniformity (by suppressing hot spots). Today, several manufacturers have commercialised this type of burners. Industrial demonstrations have been mainly validated in Asia (slab, billet or reheating furnaces, etc). These types of burners operate in the « flameless oxidation » mode. In Europe, the first demonstration appeared several years ago.

A lot of work has been done for several years to develop reliable prediction tools in order to study the different options to modify existing or develop new furnaces. Moreover, studies are in progress to evaluate the opportunity of installing such technologies in other industrial sectors (ceramics, petrochemicals, glass melting, gas turbines, etc.). Gaz de France has been particularly active on the subject for the past years. The main goal of its studies is to promote the diffusion of these new burners in France and in Europe, through a better knowledge of the phenomena inherent to this combustion mode, and by developing new design tools to ensure the installation efficiency, and the heating quality.

The main results obtained in the different following fields of action are presented in this paper.

2. TESTING OF BURNERS OPERATING IN FLAMELESS MODE

Performances of four flameless regenerative burners were compared to those obtained by using a conventional system firing. Gaz de France has carried out an experimental set-up which allows to study the influence of the main operating parameters (i.e. the furnace temperature, the thermal input, the air-fuel ratio, …) of the burners on their combustion efficiencies and NO_x emissions.

The results of the tests were obtained over several years through different studies. Globally, we showed that the flameless oxidation regime allows to reduce the NO_x emissions considerably (maximum 300 mg/m^3(n) at 3% O_2, with a furnace temperature of 1300℃ and a combus tion air temperature of 1000℃) while improving the combusti on efficiency.

We underlined the influence of the main operating parameters of these regenerative burners:

- Increase in the NO_x emissions (NO_x levels doubles from 1100℃ to 1300℃), and lower combustion efficiency (-8% with the same variation) when the furnace temperature increases,
- Strong influence of the air/fuel ratio on the NO_x emissions (NO_x levels double from 1.05 to 1.20),
- Small influence of the thermal input on the NO_x emissions in spite of a greater impact on the combustion efficiency,
- Strong influence of the percentage of exhaust gases recirculation on the combustion efficiency (+15% when recirculation ratio goes from 50% to 82%), without affecting the NO_x emissions.

The main conclusion of these tests is that several very low NO_x burners are now becoming commercially available. However, industrial references are still very rare in Europe because of a lack of industrial tools to design such installations.

3. INTERNOX PROJECT

Gaz de France initiated a project with joint funding from ARCELOR Research, Stein-Heurtey (furnaces designer) and the French Environment Agency (ADEME), to evaluate the capabilities of the HiTAC (High Temperature Air Combustion) technology. Industrial technological monitoring and detailed study of combustion in a burner of the flameless oxidation type (detailed measurements, numerical simulations, etc.) was used during the project. In addition, a validation of these tools has been conducted on a semi-industrial scale on several test cells, which were finely instrumented and put in place for this purpose. This project allowed us to develop numerical tools to design installation of such burners into a process, and industrial constraints (control, maintenance, etc.) were studied.

3.1. Experimental in-flame investigation

Within the framework of a PhD Thesis with the CORIA – laboratory (Rouen), detailed in-flame measurements (temperature, velocity, species, radiation, etc.) in stationary mode were carried out. These experimental data were used for a better knowledge of the physicochemical characteristics of this flame but also for validation of the numerical simulation of the burner.

The burner used is extrapolated from the NFK burner characterised within the project framework. It consists of two off-axis natural gas injectors set symmetrically around a central air duct. It is installed on a furnace which is instrumented to allow the measurements of global characterisation of the combustion regime, and detailed measurements in the flame. Several test-cases were studied around the reference operating conditions corresponding to operation close to real conditions:

- Thermal input = 200 kW,
- Air fuel ratio = 1.1,
- Preheated air temperature = 1000°C,
- Furnace temperature = 1300°C.

Detailed measurements were carried out. For instance, we present, on the figure 1, results of the average flame temperature measurements. We can observe weak temperature gradients and maximal temperature at 1550°C. This temperature cor responds to the thermal NO_x formation threshold temperature. In the recirculation zones, the temperature is homogeneous and close to the furnace temperature (1300°C). If one is interested in the e volution of temperature fluctuations, one notices two zones, located between the air flow and the two gas injections, where the fluctuations reach values of about 10%. Beyond, these fluctuations are very weak, and this phenomenon is one of the remarkable characteristics of this combustion mode.

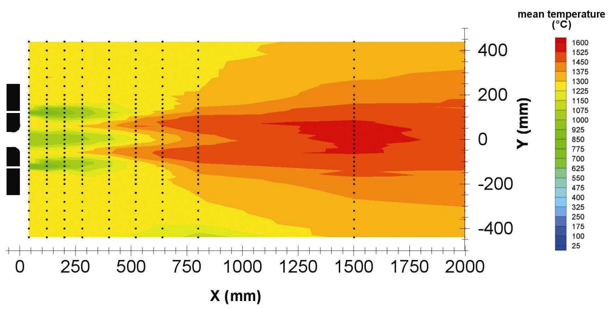

Figure 1: Mean fields of the temperatures obtained in the flame

These measurements analysis allow to study the mechanisms governing the combustion mode: flame stabilisation conditions, quantification of combustion products recirculation rate which is responsible of the species dilution, identification of the NO_x formation mechanism. As a result of the employed resources, this study constitutes an original work which allowed to provide the data still rare in the literature. In the long term, this work should lead to a better understanding of the phenomena governing flameless oxidation will help us in its application to other industrial application types.

3.2. Simulation of the burner and simplified representation

The CFD approach selected and the obtained model represents the detailed physical phenomena (flows, thermal transfer, combustion, etc.). The employed methodology is firstly to pre-select the sub-models suitable to represent the industrial burners, for then validating them starting from the detailed in-flame measurements.

The numerical simulation and experimental validation showed that it is possible to simulate a flameless oxidation burner with CFD but using standard turbulence, radiation and combustion models. The prediction of the aeraulic and thermal fields is in adequacy with the experimental data.

A detailed model is very precise but cannot be implemented to simulate an industrial scale furnace with an acceptable CPU time cost. A simplification methodology based on CFD was thus developed. In this model, the flow was simplified, and the solved equation number was reduced. This approach allows to considerably reduce the mesh size, and consequently reduce the computing time of several hours to a few minutes, while preserving a comparable accuracy. The selected strategy consists in simplifying CFD model in two stages:

- The representation of the fuel and oxidizer jets by a single equivalent jet with combustion products composition,
- The representation of the combustion zone by a volumetric source term.

The accuracy of this simplified CFD model compared to the detailed model was evaluated, and the minimal mesh size allowing to preserve an acceptable accuracy was found. These conclusions were implemented to simulate a semi-industrial scale installation in dynamic mode.

3.3. Test of the burner in a batch type furnace

3.3.1. Introduction

Before using the tools at an industrial scale, and simulating the complete installation, a preliminary validation work was conducted on a semi-industrial scale. For this, a test furnace was specially designed and manufactured by Stein-Heurtey. The NFK regenerative burners previously used was tested under the steel reheating operating conditions to reach the following objectives:

- To constitute an experimental database in order to validate the developed modelling tools,
- To evaluate the performance of the flameless-oxidation burners under an industrial furnace conditions. One is interested in particular in the heating effectiveness and quality (the NO_x emissions and the intrinsic performance have already been analysed during the characterization tests),
- To acquire a technical expertise which will enable us to ensure this technology transfer towards industrial applications.

3.3.2. Description of the test furnace

This furnace at semi-industrial scale shows the following characteristics:

- It is equipped with two NFK HRS-DL 200kW regenerative flameless oxidation burners positioned 3 meters apart face to face and in the furnace upper zone,
- It is equipped with a furnace loading/unloading system allowing to introduce a steel slab measuring 1m x 1m x 0.22m.

The tests furnace was especially designed to receive the flameless oxidation burners. This furnace originality is its capacity to receive a steel slab (see figure 2). It is thus possible to quantify the heat transfer between the source term (the combustion products), the furnace walls, and the steel slab.

For this test at semi-industrial scale, an appropriate carbon steel conventional slab was supplied by Arcelor and was instrumented with thermocouples to measure temperatures during the test.

3.3.3. Main results

The NFK HRS-DL regenerative flameless oxidation burners were implemented in semi-industrial conditions. The collected measurements allow to validate the developed numerical tools. In addition to the generation of an experimental database generation, this test programme permits to evaluate the general behaviour of the burners, i.e.:

- The NO_x emission levels are very low and in agreement with the characterisation study carried out by Gaz de France,
- The combustion efficiency is higher than 85% under the reference operating conditions, and the combustion products leaves the regenerative heat capacity at a temperature lower than 100℃, whatever the temperat ure level of the steel slab,
- The pulsed regulation mode appeared very satisfactory for the burner control and offers advantages of heating quality and control of NO_x emission control,
- The burners allows to achieve a very good thermal homogeneity along the furnace,

- When the calorific request is low, the regenerative heat capacity effectiveness falls off.

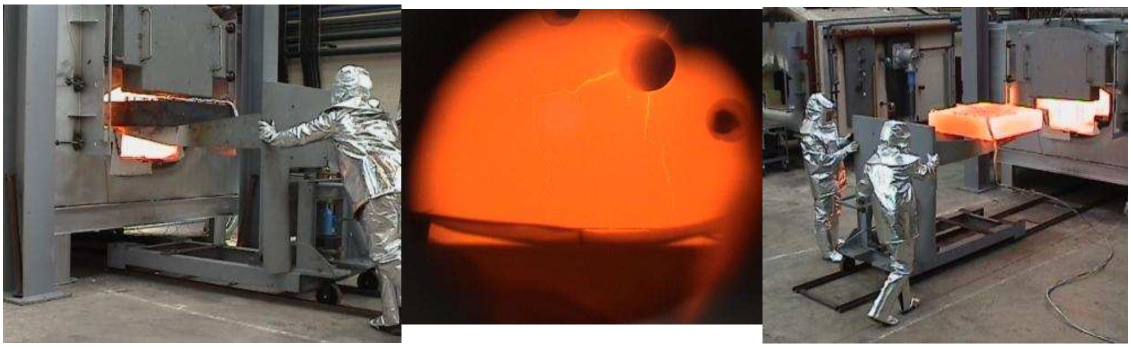

Figure 2: (a) Loading of the slab, (b) Heating of the slab by the burners, (c) Unloading of the slab

Temperature curves obtained during the long-duration test

These tests, carried out with the loading of an instrumented slab (at different temperatures and for different operating conditions) constitute an invaluable database for comprehension, validation and the definition of the tools operational limits at semi-industrial scale.

3.4. Simulation of the semi-industrial scale furnace in unsteady state

3.4.1. Introduction

We first determined the flow structure and the heat transfer nature in the furnace in steady state conditions with CFD calculations. This showed that these "high-temperature" processes are characterized by high radiative heat transfer to the load which represents more than 90% of the observed total heat exchange.

Secondly we used another approach which constitutes a global solution, similar to the zones model, focused on a study of the unsteady heat transfers. This approach, called CFD-ZONE, is used to obtain a numerical representation of the load temperature rise in a "batch" furnace.

3.4.2. Main results

The different numerical results are compared to the experimental measurements. The results concerning the load temperature are in good agreement with the experimental data, despite of the division use into very small zones. The time curves of the rise obtained numerically are in agreement with the experimental measurements (see figure 3).

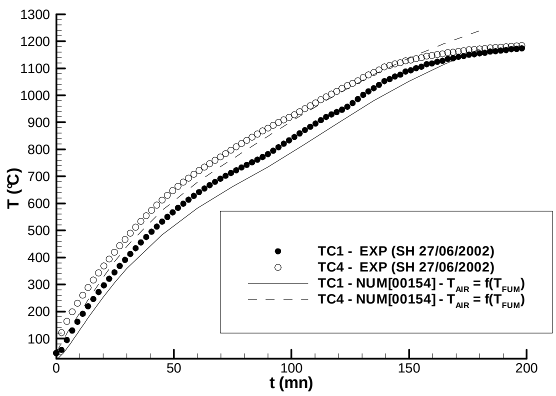

Figure 3: Experimental and numerical temperature in the slab during the time in the semi-industrial test cell

3.4.3. Conclusion

We have an operation and validated numerical tool. This tool can be used to carry out a pre-design of an industrial facility. In addition, it can allow to quantify the impact of a new burner technology use on the metallurgical furnaces or any other "high-temperature" processes. Indeed, this tool remains a generic tool allowing to estimate the dynamic heat transfer for industrial installations.

Moreover, the developed global approach takes the advantage of the software follow-up necessary for the introduction of a simulation study, and in particular of pre-treatment and post-treatment software. Although based on a CFD model, the implementation of a numerical simulation using this tool is considerably simplified compared to a conventional CFD approach.

3.5. Demonstration operation

A reheating furnace was studied to be equipped with regenerative burners in the preheated zone (increasing the capacity from 10 to 15 %). The simulation of the furnace (see figure 4) done with the tools developed in the project showed that increase of production is possible without affecting the specific consumption and without affecting NO_x emissions.

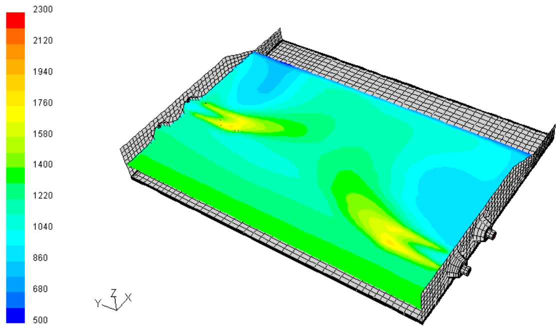

Figure 4: Temperature field on the furnace

3.6. Odyssee project

A new two years project supported by Ademe began with the same partners in December 2005 whose objective is to define an overview of the potential process in metallurgy that are concerned with the installation of flameless oxidation regenerative burners. Several studies inside the Arcelor group are made during this project.

4. A LABORATORY SCALE FACILITY TO ASSESS NOTABLY THE EFFECT OF CONFINEMENT ON FLAMELESS COMBUSTION REGIME

An experimental facility at laboratory scale has been designed and set up at CORIA (Rouen) in collaboration with Gaz de France for the study of flameless combustion mode. The unique characteristics of the facility are the possible easy change of the combustion chamber dimensions while keeping same other operating conditions. This allows one to isolate the effect of confinement ratio on the flameless combustion regime. Parametric studies of preheating air temperature and excess air ratio show results similar as the ones previously obtained at semi-industrial scale. This validates the ability of the facility to reproduce the flameless combustion mode. The aim of this study is to contribute to this understanding in order to be able to give some keys to extend the flameless combustion concept to other combustion applications.

4.1. Characteristics of the facility

The laboratory scale facility (see figure 5) has been designed in order to:

- Keep similar operating conditions as previous semi-industrial ones, notably high wall temperature,
- Be fully optically accessible for laser diagnostics,
- Enable easy change of combustion chamber dimensions for the variation of flow confinement.

These characteristics are obtained thanks to the modular configuration of the combustion chamber.

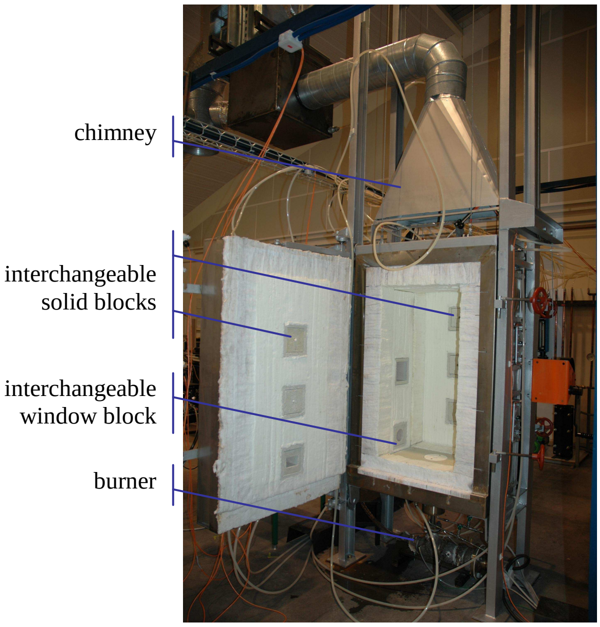

Figure 5: Global view of the combustion chamber in the low confinement configuration.

For the present study, the burner has been designed by similitude with the semi-industrial one. It consists of two off-axis natural gas injectors set either side of a central air duct. Its dimensions are calculated to have same values for the two model burners of the ratio of air and natural gas inlets velocities, and the predicted recirculation ratio calculated thanks to the Craya-Curtet theory.

4.2. Operating conditions

The laboratory facility is equipped with several measuring and controlling devices. Methane or network natural gas can be used as fuel. Inlet flowrates of gas and air are measured by volumetric flowmeters. Operation of the facility in stationary regime requires the preheating of the input air, which can be done up to 800℃ thanks to an electric heate r.

Nominal operating conditions of the laboratory facility are:

- Thermal input power: P=20 kW,
- Excess air ratio: λ=1.1,
- Air temperature: T_{air}=600℃,
- Wall temperature: T_w=1100℃,
- Low confinement configuration.

As expected in this specific "colorless" combustion mode, no visible flame can be seen in the combustion chamber (Figure 6). One observes rather evanescent fluctuating glow due to refractive index variations as during semi-industrial scale experiment, and a few soot emissions for large excess air ratio.

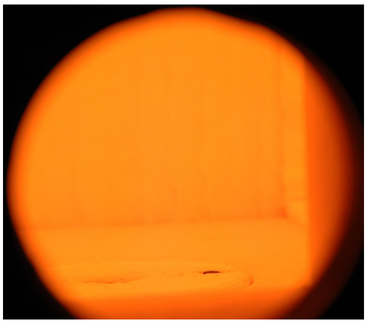

Figure 6: Photograph of the facility close to the burner exit in flameless combustion regime.

A parametric study has been performed from this reference test case by changing several parameters, notably air temperature, excess air ratio and confinement configuration. Main conclusions are that :

- Even if recirculation ratio increases with the **combustion air temperature**, which induce a larger dilution of the injected gas and air by recirculating combustion products, we observe an increase of NOx concentrations measured in flue gas. Same evolution has already been pointed out when benchmarking different regenerative burners in flameless combustion mode. In the present case, NOx concentrations vary from 14 mg/m^3 @ 3% O_2 at ambient air temperature to 65 mg/m^3 @ 3% O_2 at T_{air} = 600℃. As thermal input power is kept constant i n the present experiment, total input power increases when preheating air. This explains the increase of wall and flue gas temperature, and may induce also an increase of local maximum temperature in the flame, which then would be responsible of an increase of NO formation via the thermal route.
- Increase of **excess air ratio** in the considered range (1.1 to 2.2) does not induce significant increase of air jet momentum flux. Recirculation ratio is then quasi constant in this range. No drastic change in the global aerodynamic of the flow in the combustion chamber is then expected when varying excess air ratio. Variation of excess air ratio from the reference test-case induces a large increase of NOx emissions. NOx concentrations vary quasi linearly from 50 mg/m^3 @ 3% O_2 at n = 1.1 up to 240 mg/m^3 @ 3% O_2 at n = 2.2. For large excess air ratio over the stoichiometry, recirculating products contain significant amount of oxygen, which has not reacted with fuel. Then dilution of injected air and gas is not only done with inert combustion products but also with a significant part of oxygen. The latter increases again the local oxygen concentration. Operating conditions move away from MILD combustion mode to a more standard one, which because of the high local temperature, induces larger NOx emissions from thermal NO formation.
- **Variation of combustion chamber dimensions** from the low confinement configuration to the high confinement one has a small effect on flue gas temperature, which slightly increases because of a higher heat release density. High confinement of the flow for same burner operating conditions decreases naturally the value of the recirculation ratio by approximately 80% in our case. No significant change of NO_x emissions is observed for the reference test case, whereas a slight increase is noticed for λ=1.6. In these two cases, air preheating ensures large value of recirculation ratio even in high confinement configuration. So the effect of dilution by recirculating products could be still important and may explain the weak changes in NOx emissions.

5. MODELISATION OF FLAMELESS COMBUSTION WITH A NETWORK OF IDEAL REACTORS AND DETAILED CHEMICAL KINETICS

The present work introduces an original approach to predict the NOx emissions stemming from flameless oxidation in a furnace with a network of ideal reactors and detailed kinetics of combustion. It is based on a simplified flow model and a detailed chemical kinetics model.

- Flameless oxidation principle, with strong recirculations and diluted combustion, can be assimilated to ideal reactors such as perfectly stirred or plug flow ones. Thus, the flow model consists of a simple network of this kind of reactors. It is feeded with residence time distribution (RTD) measurements.
- The chemical kinetics model, GDF-Kin®3.0, developed by Gaz de France. It is a detailed kinetic mechanism specifically designed to calculate natural gas combustion in various conditions. It includes the chemistry of alkanes encountered in natural gas (methane, ethane, propane, butane, pentane and hexane) as well as their interactions with NOx chemistry. This model, coupled with heat losses, allows to precisely predict pollutants emissions and especially NOx emissions for each reactor of the global previously determined network.

5.1. Experimental apparatus and procedure

In order to build the flow model which consists of a network of ideal reactors, radiotracer experiments have been carried out to get RTD of the flow in the furnace. Thus, for each case, at the chimney was collected a response to a pulse injection of radiotracer both at gas input and air input (example on figure 7).

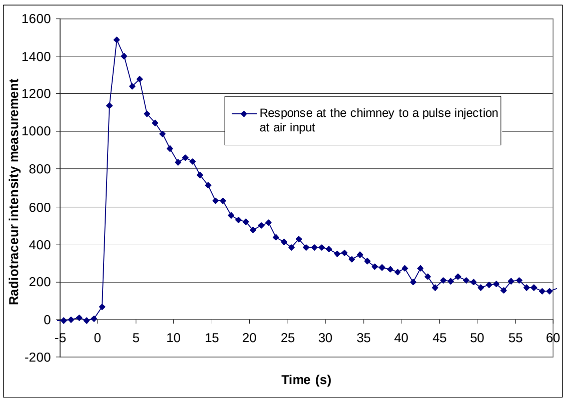

Figure 7: Radiotracer curve at the chimney, response to a pulse injection at the air input for case n°3

5.2. Modelling and simulations

Flameless oxidation modelling has been built according to the following steps:

The flow model: the network of ideal reactors was found by comparing theoretical RTD with experimental RTD. These comparisons have lead us to a simple network.

A global validation of the model has been performed by comparison of the calculated NO_x emissions and measured emissions. Results of this comparison are presented below.

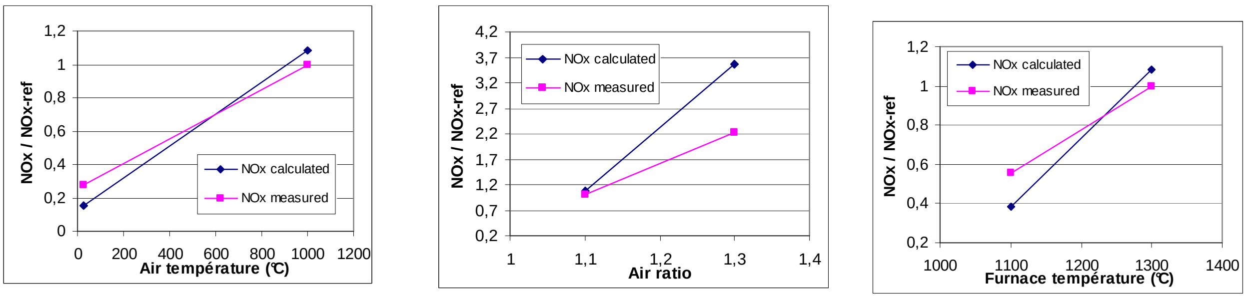

Figure 8: (a) Influence of air temperature ; (b) Influence of air ratio; (c) Influence of furnace temperature

The levels and the main trends are well reproduced. These results allow us to carry on with this particular approach in order to simulate combustion in flameless mode.

5.3. Conclusion

Simulation of flameless oxidation with a network of ideal reactors approach gives satisfactory results. This original approach makes possible the use of a detailed chemical kinetic mechanism of natural gas oxidation. While freeing oneself from using a CFD modelling, one can, in a fast and very flexible way, predict chemical phenomena such as NOx emissions.

This approach is particularly adapted to flameless oxidation due to the quite diluted combustion and stirred flow. Thus, the computational package presented here constitutes a useful and innovative tool to characterise flow pattern, associated heat transfers and detailed chemistry to better understand the flameless oxidation technology in industrial processes.

6. CONCLUSIONS

Gaz de France's Research and Development Division has been very much involved in flameless combustion for a long time. The better understanding of the combustion phenomena of the "flameless-oxidation" mode but also the design tools developed within the different activities presented in this paper and another coming results, will enable to implement this technology and give guarantees on the performances of future installations like on the products heating quality or productivity.

Even though the first industrial applications mainly concern today the metallurgy area, this new combustion system can be used, with short or medium term, in other industrial sectors such as, glass-making, gas turbines, waste treatment, petrochemicals or industrial boilers. At present, Gaz de France is involved in several projects studying this subject.

ACKNOWLEDGEMENTS

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