DEVELOPMENT OF LABORATORIAL LPP COMBUSTION CHAMBER FOR NO_x EMISSION CHARACTERIZATION IN GAS TURBINES

Armando Caldeira-Pires

Dept of Mechanical Engineering Faculty of Engineering – University of Brasilia 70910-900 Brasilia DF Brazil armandcp@unb.br

Himilsys Hernández González

Dept of Mechanical Engineering Faculty of Engineering – University of Brasilia 70910-900 Brasilia DF Brazil

Hugo Faria de Moura

Dept of Mechanical Engineering Faculty of Engineering – University of Brasilia 70910-900 Brasilia DF Brazil

Abstract. This work describes an on-going experimental study in a laboratory model of a LPP (lean premixed pre-vaporization) combustion chamber, aiming at the improvement of the knowledge of the chemical kinetics formation of thermal and prompt NO. The technological contribution of this project lies on a advanced 3D monitoring non-intrusive system, based on CCD commercial camera, and on computational algorithm to reconstruct the three-dimensional field of the emissions sources of interest, namely chemical species that emit preferentially in the UV spectrum band, namely the visualization of thermal and prompt NO production areas. The experimental model is constituted by two main parts, where optical access is provided by means of a glass tube in the pre-chamber and a tube of UV-quartz in the combustion chamber. The results to be reached at the end of this project will allow an optimization of the combustion process, through the characterization of the influence of the burner geometric parameters in the processes of turbulent mixture and fossil fuel oxidation, and the inimization of NOx formation, with the identification of the main kinetic mechanisms oxides nitrogen formation, between thermal and prompt mechanisms in gas turbine combustion.

Keywords gas turbine, LPP, NOx, optical methods, experimental characterization

1. Introduction

Combustion is a source of energy very used in almost all human economic activities. Moreover, roughly in all of these systems the combustion process occurs after a phase of intense turbulent mixture, allowing a homogeneous blend of the reactants. These processes can be found inside the combustion chambers of internal combustion engines, industrial burners, aeronautic turbines and thermal power plant gas turbines.

During the last two decades new legislation regarding more restrictive pollutants emissions have driven gas turbines manufacturers to design new combustion chambers that emit less nitrogen oxides (NOx) and less carbon monoxide (CO). This pushes the development of two innovative technologies, namely RQL (Rich Quench Lean) and LPP (Lean Premixed Prevaporized) technologies, both making use of the present knowledge on the effect of fuel/air equivalence ratio on NOx formation.

Due to these facts, and in order to maintain their competitiveness, and therefore their economic sustainability, gas turbine producers, as well as electric energy providers based on thermal power plants, have carried on extensive research work to improve their knowledge on those reactive phenomena, such as the combustion processes, occurring inside those combustors, to build new and efficient combustion chambers. Figure 1 presents the diagram of two practical LPP combustors models, respectively designed by SNECMA and by Rolls-Royce.

This work is part of a two year project (2002-2003), accomplished by a partnership between Eletronorte and University of Brasilia, aiming at characterizing the specificities of reaction and flow in thermal power plant gas turbine LPP burner, namely mixing zones, diffusion and premixed flames with various equivalence ratio. The full project encompasses non-reacting and reacting flow modeling through CFD commercial codes to assess mixture and reaction zones characteristics; combustion chemical kinetics modeling to improve knowledge on the relative importance of thermal and prompt NOx formation mechanism inside LPP combustion chamber; and experimental characterization of non-reacting and reacting zones making use of advanced imaging and tomographic reconstruction techniques to depict the influence of flow, combustor geometry and equivalence ratio on the flame.

The results presented in this paper addresses the last topic, and the rest of the paper describes NOx formation mechanisms, addressing those specific issues related with lean premixed burn, and presents in detail the experimental rig and the diagnostic technique to be used during this research. The project of the combustion chamber improves that formerly reported by Anacleto and Heitor (1997).



Figure 1. Schematic Diagram of Practical LPP Models (adapted from Legier, 2001): a) designed by SNECMA

b) designed by Rolls -Royce

2. Theoretical Concepts

Low fuel/air equivalence ratio, staged combustion, addition of inert, are methods for reducing emissions of oxides of nitrogen. There remains, however, a lack of detailed understanding of the reasons that these techniques are effective. Because of this deficiency, it is difficult to optimize combustors for achieving best performance with minimum emissions. This part of the work intends to structure the present knowledge of the fundamental factors that determine the effects of the low-NOx methods listed above, thereby adding rational approaches to improvements in design of combustors.

2.1. Nitrogen Oxides

Nitrogen oxides, or NOx, is the generic term for a group of highly reactive gases, all of which contain nitrogen and oxygen in varying amounts.

Nitrogen oxides form when fuel (hydrocarbon or hydrogen, as well) is burned at high temperatures, as in combustion processes. The primary sources of NOx are therefore motor vehicles, electric utilities, and other combustion based industrial, commercial, and residential sources.

NOx causes a wide variety of health and environmental impacts because of various compounds and derivatives in the family of nitrogen oxides, including nitrogen dioxide, nitric acid, nitrous oxide, nitrates, and nitric oxide.

Since 1970, EPA has tracked emissions of the six principal air pollutants - carbon monoxide, lead, nitrogen oxides, particulate matter, sulfur dioxide, and volatile organic compounds. Emissions of all of these pollutants have decreased significantly except for NOx which has increased approximately 10 percent over this period.

2.2. NOx Chemical Kinetics Mechanisms

Most of the nitric oxide (NO) formed in combustion subsequently oxidizes to NO_2 . For this reason it is customary to lump NO and NO_2 together and express results in terms of oxides of nitrogen (NO,), rather than NO. It can be produced by four different mechanisms, Thermal NO, Prompt NO, Fuel NO and Nitrous Oxide Mechanism, as follows:

2.2.1. Thermal NO

This is produced by the oxidation of atmospheric nitrogen in high temperature regions of the flame and in the post flame gases. The process is endothermic and it proceeds at a significant rate only at temperatures above around 1850 K. Most of the proposed reaction schemes for thermal NO utilize the extended Zeldovich mechanism:

| $O_2 = 2O$ | (1) |
|--------------------|-----|
| $N_2 + O = NO + N$ | (2) |
| $N+O_2 = NO+O$ | (3) |
| N+OH = NO+H | (4) |

NO formation is found to peak on the fuel-lean side of stoichiometric. This is a consequence of the competition between fuel and nitrogen for the available oxygen. Although the combustion temperature is higher on the slightly rich side of stoichiometric, the available oxygen is then consumed preferentially by the fuel.

NO emissions are very dependent on flame temperature, an increase in inlet air temperature would be expected to produce a significant increase in NO, and this is confirmed by the results of Rink and Lefebvre (1989). Moreover, combustor residence time can also influence NOx emissions. NOx emissions increase with an increase in residence time, except for very lean mixtures ($\phi \equiv 0.4$), for which the rate of formation is so low that it becomes fairly insensitive to time. Similar results showing the insensitivity of NOx formation to residence time in lean premixed combustion have been obtained by Leonard and Stegmaier (1993) and Rizk and Mongia (1993).

All these findings have important practical implications to the design of lean premixed combustors:

- Thermal NO formation is controlled largely by flame temperature;
- Little NO is formed at temperatures below around 1850 K;
- For conditions typical of those encountered in conventional gas turbine combustors (high temperatures for only a few milliseconds), NO increases linearly with time but does not attain its equilibrium value;

(5)

- For very lean premixed combustors ($\phi < 0.5$) NO formation is largely independent of residence time.

2.2.2. Prompt NO

Under certain conditions, NO is found very early in the flame region, which is in conflict with the idea of a kinetically-controlled process. According to Nicol et al. (1992) the initiating reaction is

 $N_2 + CH = HCN + N$

The balance of the prompt NO mechanism involves the oxidation of the HCN molecules and N atoms. Under leanpremixed conditions, the HCN oxidizes to NO mainly by a sequence of reactions involving HCN ? CN ? NCO ? NO. The N atom reacts mainly by the second Zeldovich reaction.

The influence of pressure is of special interest and importance because prompt NO can be a significant contributor to the NO emissions produced in lean premix (LPM) combustion. Unfortunately, few data are available on this effect. Fennimore's (1971) pioneering study of prompt NO in ethylene-air flames over a range of pressures from 1 to 3 atm concluded that prompt NO α P^{0,5}. Later work by Heberling (1976) over a much wider range of pressures from 0.1 to 1.8 MPa showed that prompt NO was independent of pressure. Altermark and Knauber (1987) also concluded that NOx is independent of pressure for equivalence ratios below 0,6.

2.2.3. Fuel NO

Light distillate fuels contain less than 0.06 percent of organically-bonded nitrogen (usually known as fuel-bound nitrogen), but the heavy distillates may contain as much as 1.8 percent. During combustion, some of this nitrogen reacts to form the so-called "fuel NOx" The fraction of nitrogen undergoing this change increases only slowly with increasing flame temperature. As far as gaseous fuels are concerned, natural gases contain little or no fuel-bound nitrogen, but some is found in certain process and low-Btu gases. Depending on the degree of nitrogen conversion, fuel NO can represent a considerable proportion of the total NO.

2.2.4. Nitrous oxide mechanism

| According to Nicol et al. (1992) this mechanism is initiated by the reaction | |
|--|-----|
| $N_2 + O = N_2O$ | (6) |
| and the N2O (nitrous oxide) formed is then oxidized to NO mainly by the reaction | |
| $N_2O+O = NO+NO$ | (7) |
| but also by the reactions | |
| $N_2O+H = NO+NH$ | (8) |
| $N_2O + CO = NO + NCO$ | (9) |
| | |

Nicol et al. (1992) analytically examined the relative contributions of the various mechanisms discussed above to the total NO, emissions produced by a lean-premixed combustor burning methane fuel, for which the fuel NO is zero. The results of their study show that at relatively high temperatures of around 1900 K, and equivalence ratios of around 0.8, the contributions are about 60 percent thermal, 10 percent nitrous oxide, and 30 percent prompt. With reductions in temperature and equivalence ratio, the contributions made by nitrous oxide and prompt NO increase significantly until, at a temperature of 1500 K and an equivalence ratio of around 0.6, the relative contributions to the total NO, emissions become 5 percent thermal, 30 percent nitrous oxide, and 65 percent prompt. At the lowest equivalence ratios (ϕ = 0.5 to 0.6), the major source of NO, is that formed by the nitrous oxide mechanism. These results clearly have great importance to the design of ultra-low NO, lean-premixed combustors.

2.3. LPP Combustion

A common feature of all the dry low NOx combustors is that positive efforts are made to eliminate local regions of high temperature within the flame by mixing the fuel and air upstream of the combustion zone. The lean, premix, prevaporize (LPP) concept represents the ultimate in this regard. Its underlying principle is to supply the combustion zone with a completely homogeneous mixture of fuel and air, and then to operate the combustion zone at an

equivalence ratio which is very close to the lean blowout limit. The smaller the margin between stable combustion and flame blowout, the lower will be the output of NOx.

A typical LPP combustor can be divided into three main regions. The first region is for fuel injection, fuel vaporization, and fuel-air mixing. Its function is to achieve complete evaporation and complete mixing of fuel and air before combustion. By eliminating droplet combustion and supplying the combustion zone with a homogeneous mixture of low equivalence ratio, the combustion process proceeds at a uniformly low temperature (< 1900 K) and very little NO (below 50ppm), is formed. In the second region the flame is stabilized by the creation of one or more recirculation zones. Combustion is completed in this region and the resulting products then flow into region three which may comprise a fairly conventional dilution zone.

A useful by-product of LPP combustion is that it is essentially free from carbon formation, especially when gaseous fuels are used, in which case the description "lean premixed" or "LPM" is more appropriate. The absence of carbon not only eliminates soot emissions but also greatly reduces the amount of heat transferred to the liner walls by radiation, thereby reducing the amount of air needed for liner wall cooling. This is an important consideration because it means that more air is made available for lowering the temperature of the combustion zone and improving the combustor pattern factor.

Another important advantage of LPP systems is that for flames in which the temperature nowhere exceeds 1900 K the amount of NO formed does not increase with increase in residence time. This means that LPP systems can be designed with long residence times to achieve low CO and UHC, while maintaining low NO levels. This finding is especially significant for industrial engines, where size is less important than for aero engines. As noted above, this approach leads to an LPM combustor volume which is approximately twice that of a conventional combustor.

The main problem with the LPP concept is that the long time required for fuel evaporation and fuel-air premixing upstream of the combustion zone may result in the occurrence of auto-ignition at the high inlet air temperatures and pressures associated with operation at high power settings. Another problem which is associated with all well-mixed combustion systems is that of acoustic resonance which occurs when the combustion process becomes coupled with the acoustics of the combustor (1998).

3. Experimental LPP Combustion Chamber Development

Figure 2 shows the schematic diagram of the physical model being developed within this study, which encompassed a premixing chamber and the combustion chamber, itself. A bluff-body is positioned at the entrance of the combustion chamber to stabilize the flame. A detailed description of this installation is provided elsewhere (Anacleto and Heitor, 1997), and Table 1 depicts the flow condition range to be analyzed in the scope of this project.



Figure 2. Schematic Diagram of the LPP Laboratory Model.

In this experimental work, LPG (C_3 and C_4 hydrocarbon) is selected for the fuel, mainly because is the easiest low molecular weight hydrocarbon commercially available. The fuel and the air were used in atmospheric conditions.

Table 1. Flow Conditions Range to be used during the Experiments

| Equivalence Ratio | Mass Flow _{Fuel} .x 10^{-3} (kg/s) | Mass Flow _{Air} x 10 ⁻³ (kg/s) | Re (calculated) |
|-------------------|---|--|-----------------|
| 0,6 | 1,1405 | 29,415 | 31860 |
| 0,8 | 1,5206 | 29,415 | 32280 |
| 1 | 1,9008 | 29,415 | 32700 |
| 1,2 | 2,2810 | 29,415 | 33120 |

This installation has been modeled in a commercial CFD code, aiming at allowing further studies on flow conditions and geometry effects on the fuel burning. Figure 3 displays the tri-dimensional diagram of this system, showing its main geometric lines and its rendered image as well.





4. Diagnostic Methods : Chemiluminescence Image Acquisition

A 10bits CCD digital camera is placed perpendicular to the UV-quartz (up to 60% transmittance at 220nm) section of the combustor, obtaining radial views of the flame. The video output signals were connected to a frame grabber, and used for the acquisition of independent monochromatic video signals. A commercial 50mm UV lens (f = 1.4) allowed a cone collection of light of 20°, minimizing the distortion due to wide-angle imaging.

Visible interference filters (centered at 226nm for NO free radical imaging, 308nm for OH, 355nm for NH and 388nm for CN, with transmittance ranging from 15 to 50%) are used for acquisition of monochromatic flame images. A dedicated software library was used in order to control the frame grabber. Tomographic reconstruction software was purpose built, performing images ratio calculation as well, based on the description of Correia (2001).

The flame images acquisition procedure consisted on obtaining time-averaged images for the two wavelengths, for a single projection, as the average flame characteristics are axi-symmetric.

Figure 4 shows the tomographic reconstruction process applied on a propane C_2 flame image, depicting reaction and recirculation zones within the combustion chamber, as reported by Caldeira-Pires (1999).



Figure 4 Mean Projection and Reconstructed Image of Chemiluminescence emission (adapted from Caldeira-Pires, 1999)

a) CH Free Radical Chemiluminescence Projection (prompt NO images are expected to follow the same patterns, both intensity and position)

b) Tomographic Reconstruction Process

c) Free Radical Chemiluminescence Reconstructed Image.

Due to low emission levels for NO free radical emission imaging (low quantity of emitting sources * low light emitted from each source * low transmittance at 226nm of filters and lens) it is added to the optical system a 2-MCP intensifier enabling up to 10^6 gain increase.

Thus, the whole optical system comprises:

- UV-quartz combustion chamber, ~60% transmittance at 220nm;
- o Interferometric filter centered at 226nm ±6nm, ~15% transmittance at 225nm;
- o UV-lens (11 elements), transmittance below 20% at 200nm;
- o 2-MCP intensifier, 65mA/W spectral sensitivity at 200nm on the incoming element;
- o 10-bit digital camera.

The optical path is depicted by Fig. 5.



Figure 5 Schematic diagram of the imaging elements on the optical path

5. Summary

In this paper we point to the NOx emission characterization as a crucial parameter whose meaningful prediction is a prerequisite for accurate calculations of the several chemical kinetics mechanisms of oxide of nitrogen production in premixed combustion. We present the first steps towards the establishment of an experimental installation to assess NOx formation characteristics inside a LPP combustion chamber, namely the zones of formation of prompt and thermal NOx.

A technique to visualize the time -averaged and short-exposure chemiluminescence images of the main intermediary species within the NO production chain reaction will be used, namely to characterize the locus of emission of the following free-radicals: C₂, CH. CN, NH, OH and NO. Due to low levels of light available, a specific optical system is designed, encompassing narrow-band interferometric filters, a UV-lens and a 2MCP intensifier. These optical components will allow the emitted light acquisition through a high resolution 10-bit camera.

As part of a bigger project, the results to be obtained will allow characterizing the specificities of reaction and flow in typical gas turbine LPP burner. Moreover, they will act as a base for reacting flow CFD modeling evaluation, as well as will highlight the relative importance of thermal and prompt NOx formation mechanism inside LPP combustion chamber.

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