

## REYNOLDS NUMBER AND EQUIVALENCE RATIO EFFECTS ON NO<sub>x</sub> EMISSION ON NON PREMIXED TURBULENT FLAMES

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**Abstract.** *The impact of Nitric Oxide emissions from combustion processes on the environment and public health has prompted strict regulations in order to reduce the emission of these pollutants. The present work investigates the effect of the flow dynamics on NO<sub>x</sub> formation and analyzes the spatial distribution of the chemical species in different flame positions. The flame considered in this work is a non-premixed flame anchored by a pilot flame. The flow dynamics is varied through the Reynolds number and the global equivalence ratio. The techniques considered to investigate the NO<sub>x</sub> formation were the gas composition analysis and temperature measurements. Natural gas is used as fuel in a Delft type burner. Based on the experimental results it was possible to understand the behavior of NO concentration when the Reynolds number is increased corroborating with the hypothesis that the NO<sub>x</sub> formation is reduced with the flow velocity increase due to the reduction on all time scales, including the residence time. The gas composition analysis was done with a stainless steel cooled probe, while the temperature measurements was done with a thermocouple Pt/PtRh13%. The experimental facility considered a well known burner that allows a detailed flame visualization in non-premixed flames, and a positioning system for the gas probe and thermocouples that traveled both in the radial and longitudinal directions. The Reynolds number was varied through the fuel velocity and three equivalence ratios were considered, poor, stoichiometric and rich. The results show that for flames with equivalence ratio of 1.3 the EINO<sub>x</sub> is greater than for poor and stoichiometric global equivalence ratio for any longitudinal position, which confirms the data available in the literature. For any global equivalence ratio at any longitudinal position the EINO<sub>x</sub> is reduced with the increase in the Reynolds number. These results could be related to the flame residence time. Reducing the residence time the time available for NO<sub>x</sub> reaction is also reduced. The temperature measurements indicate that the instant mechanism is predominant over the thermal mechanism since the average temperatures are relatively low remaining always below 1700K.*

**Keywords:** *NO<sub>x</sub> emission, diffusive flames, Delft burner*

## 1. INTRODUCTION

The fire plays an important role in human development since early ages. Nevertheless, combustion is a phenomenon whose description is still an open issue despite the progress achieved in the last century.

The impact of Nitric Oxide (NO<sub>x</sub>) emissions from combustion processes on the environment and public health has prompted strict regulations in order to reduce the emission of these pollutants. An extensive effort have been carried in order to understand the NO<sub>x</sub> formation mechanism and for the development of technologies for its reduction. The main questions that remain to be answered are related to the interactions between turbulence and the chemistry of Nox

formation (Meunier et al, 1998). No direct correlation between turbulence and NO<sub>x</sub> emission has been formally established.

The combustion of fuels that do not have nitrogen compounds, as for example natural gas, there are three routes or mechanisms for production of NO: The thermal mechanism or Zel'dovich, the prompt mechanism, or Fenimore and the intermediate N<sub>2</sub>O mechanism. In hydrocarbon combustion the first two mechanisms are responsible for most of the NO formation. While for non-hydrocarbon systems, such as H<sub>2</sub>O-air or H<sub>2</sub>-CO-air, generally only the thermal mechanism is the active one.

Most experimental and theoretical works on emissions index from turbulent jet diffusive flames have been related to the jet primary variables, such as: jet exit velocity, burner diameter, fuel chemical composition, Reynolds number and Froude number at the jet exit (Bilger e Beck, (1975); Drake et al. (1987); Turns e Lovett (1989); Driscoll et al. (1992); Turns (1995) e Caldeira Pires et al., 1997). Meunier et. al., (1998) studied NO<sub>x</sub> emissions in propane diffusive flames including experimental measurements and numerical simulations. They concluded that the EINO<sub>x</sub> results are proportional to the reaction products residence time in high temperature zones.

Santos e Costa (2005) studied diffusive flames burning propane, methane and ethylene. Three different burner diameters were considered with different exit jet velocities. Their main conclusions were: i) the transition is controlled by the momentum are related to the type of hydrocarbon; ii) the propane and ethylene flames results indicate that hydrocarbons prone to soot formation are critical for NO<sub>x</sub> emission; iii) Data from ethylene flames show that radiation effects are important when building correlations for NO<sub>x</sub> emission.

Caldeira Pires et al., 1997 presented an analysis related to the formation of NO<sub>x</sub> based on experimental results. They considered a propane diffusive flame including Zel'dovich (thermal), intermediary Nitrose oxide and the Fenimore (prompt) mechanism for different flame locations at different Reynolds numbers. They found that, at similar longitudinal locations, NO<sub>x</sub> concentration were higher for lower Reynolds numbers, corresponding to lower velocities and higher residence times. The higher NO<sub>x</sub> concentrations were observed in regions of rich flame.

Caldeira Pires et al. (2000), analysed the velocity, temperature and concentration of chemical species in different flame positions as a function of the Reynolds number in propane flames. The results were similar to their previous results (Caldeira Pires e. al., 1997). They found that NO<sub>x</sub> concentration increases with the reduction of the Reynolds number due to the increase in the residence time. They also found that the highest NO<sub>x</sub> production rates were located close to the positions of maximum temperatures. The temperature seems to be the controlling factor for the thermal NO production (de Soete, 1988), Therefore, lowering the temperature is the main goal as NO reduction strategy.

The objective of the present work is to understand the spatial distribution of NO<sub>x</sub> in a non premixed turbulent flame of natural gas, burning at atmospheric pressure. The analysis is based on the variation of the flow dynamics through the variation of Reynolds number and global equivalence ratio. The results allow conclusions to be drawn regarding the reduction of NO<sub>x</sub> formation with velocity increase due to the reduction of the characteristic times (including the residence time). In the open literature there are results indicating both an increase and a reduction of NO<sub>x</sub> concentration with Reynolds number increase (Drake et al., 1987, Turns e Lovett, 1989, Caldeira Pires et al., 1997).

## 2. EXPERIMENTAL SETUP AND METODOLOGY

The present work considers the experimental evaluation of the distribution of chemical species in different flame positions for different flow dynamics. The flame is a non-premixed flame, anchored to the burner by 12 small pilot flames. Gas composition and temperature measurements are used. The experimental set up is composed of the following items: natural gas feed line, flow control panel, burner, positioning system, sampling probe, sample conditioning system, gas analysers, data acquisition system, thermocouple and optic system. The experimental setup is described at Martins et al (2005).

The fuel oxidizer mass ratio for the equivalence ratio,  $\Phi$ , was calculated using the chemical equilibrium code "Teqworks-Thermochemical Information and Equilibrium Calculations". The test bench is located on the National Institute of Spatial Research (INPE) Propulsion and Combustion Associated Laboratory (LCP), in Cachoeira Paulista, São Paulo, Br.

Burner based on the principals layed by Starner e Bilger (1985) is a coaxial jet type which could be tough more complex than jet simple and more simple than swirl or bluff body burner, had been characterized in the Technology University of Delft – TUDelft (De Vries, 1994). The burner consists of two coaxial ducts, the external one for air with a diameter of 30 mm and the internal for natural gas with a diameter of 6 mm. The pilot flames are a acetilene/hydrogen and air used to stabilize the main flame at the burner exit.

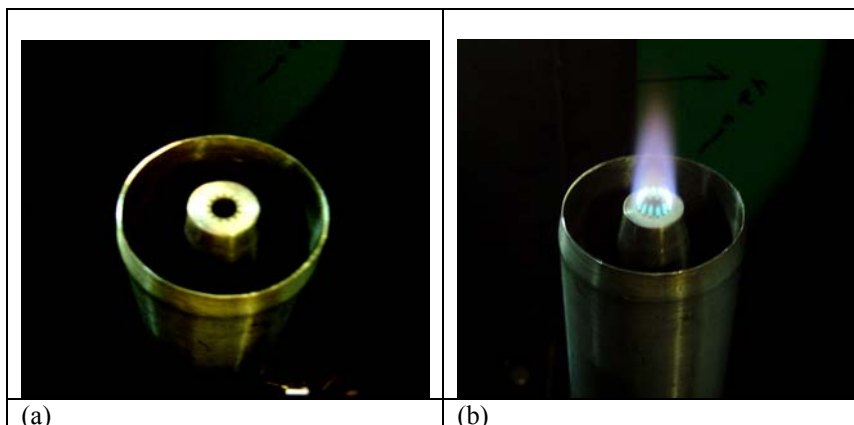


Figure 1 – (a) Pilot flames project detail; (b) C<sub>2</sub>H<sub>2</sub>/H<sub>2</sub> pilot flame.

The positioning system consists of a vertical linear scale and of a metal bar that allow measurements of 1/10 of mm in the radial and axial directions. The gas sampling in the flame was taken by a cooled probe made of stainless steel with 1 mm diameter and a suction flow rate up to 1 l/min in order to avoid disturbing the jet flow. Rosemount Analytical gas analysers were utilised. The average sampling of 1000 points were sampled after 4 minutes and followed to system acquisition which utilized LabVIEW software. Preliminary tests were performed to assure the reliability of the data sampled. This tests considered the alignment of the CCD camera, gas-leak detection, probe reliability test, flow rate measurement calibration, calibration of the analysers and test of response time.

The methodology used considers the variation of the flow dynamics and observation of the spatial distribution of chemical species of interest (CO, CO<sub>2</sub>, O<sub>2</sub> and NO<sub>x</sub>) and mass flow rates. Three different radial positions in the flame were considered. The flow dynamics was varied changing the Reynolds number of the fuel flow and the global equivalence ratio. Three Reynolds numbers were considered: 5400, 7300 and 9500. The global equivalence ratio are pour, stoichiometric and rich in fuel,  $\Phi = 0.7, 1.0$  and  $1.3$ . The Reynolds number is based on viscosity and density measured at cold temperature, with length scale taken as the diameter  $d_0$  at the burner exit and average jet velocity  $U_0$  at the nozzle exit.

The following techniques were used to measure the properties considered in the analysis: for the gas concentration analysis a cooled probe was used and the temperature measurements were done with a thin wire thermocouple (Pt/PtRh13%). Chemical species concentration was determined on a dry base.

### 3. RESULTS AND DISCUSSION

The terminology equivalence ratio and pour or rich mixtures should be understood on a restricted sense due to the fact that the flame is a free jet in air and the adopted values correspond to fuel/air ratios at the burner exit. Is very common in literature term global equivalence ratio when non premixed flame is treated, it is not true but is considered acceptable. Here the authors considered the term equivalence global.

In the coaxial jet configuration, the jet velocity decays in the streamwise direction and air is entrained from the surrounding ambient. The resulting jet topology has a conic shape and there is a boundary layer type similarity solution for the jet far from the nozzle exit.

Three different fuel jet velocities were taken, as well as three different mass fuel/air ratios. Concentration measurements were taken in three different axial longitudinal positions. At each longitudinal position, three radial positions where considered, resulting in 27 radial measurements. Table 1 presents the test conditions considered. The subscript  $J1$  and  $J2$  correspond to the fuel jet and the oxidizer jet respectively.

Table 1 – Test Conditions

Fuel		Air					
		$\Phi 0,7$		$\Phi 1,0$		$\Phi 1,3$	
$V_{j1}$ (m/s)	$Re_{j1}$	$V_{j2}$ (m/s)	$Re_{j2}$	$V_{j2}$ (m/s)	$Re_{j2}$	$V_{j2}$ (m/s)	$Re_{j2}$
15,3	5400	8,27	15635	5,79	10942	4,45	8416
20,7	7300	11,19	21158	7,83	14807	6,02	11389
27,0	9500	14,56	27529	10,19	19265	7,84	14817

### 3.1 Flame length

The main characteristic of a non-premixed flame is its shape, particularly its height. Figure 2 from Kuo (1986) show three distinct regions for a flame, the first for a laminar flow where the flame height increases linearly with the jet nozzle exit velocity. The second region is a transition region which is followed by a fully developed turbulent flow region where the flame height is independent from the nozzle exit velocity, or Reynolds number for that matter.

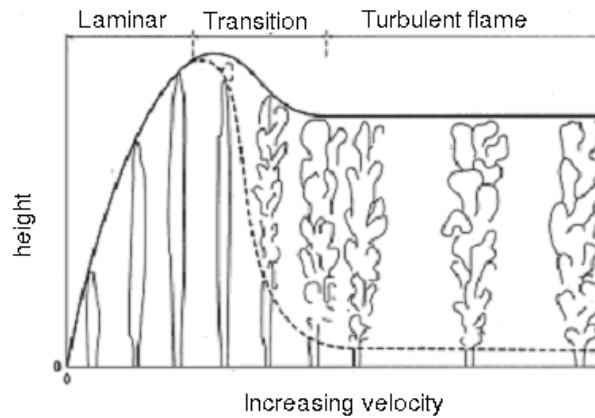


Figure 2 – Flame height variation with nozzle exit velocity (Kuo, 1986).

The flame height is an indication of how efficiently air is been entrained in the flame region. The higher the amount of air entrained the faster fuel is burned. Figure 3 shows that increasing the co-axial air velocity the flame thickness is reduced, but the flame height remains approximately the same. That may be explained by the fact that increasing the turbulence levels through higher external jet velocities the contact between reactants is increased, leading to a reduction on the flame boundary at the radial direction.

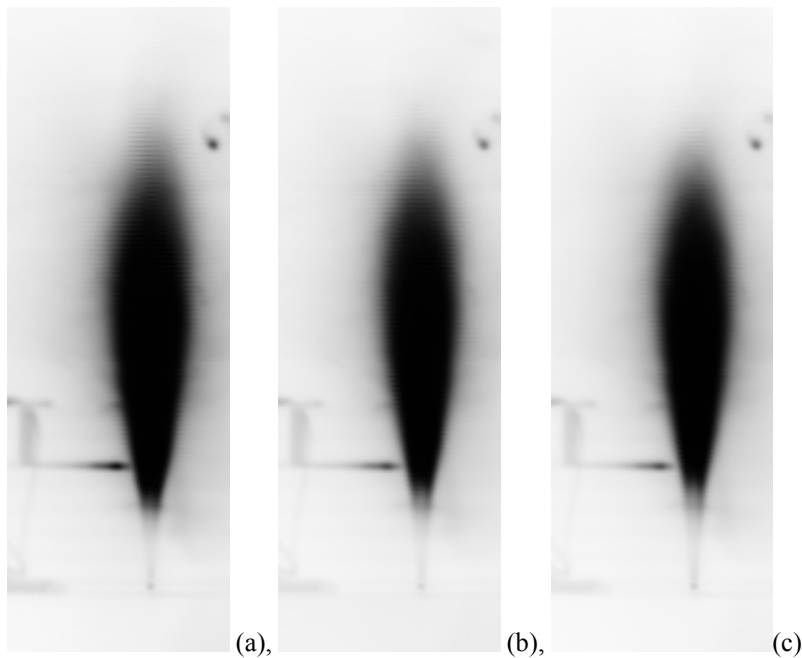


Figure 3 – Flame shape variation with external air jet velocity: (a): 4.45 (b) 5.79 e (c) 8.27 m/s, with Constant fuel jet velocity (15.3 m/s).

### 3.2 NO<sub>x</sub> Concentration

Table 2 presents the range of value found for the concentration of NO<sub>x</sub> in ppm.

Table 2 – Range of NO<sub>x</sub> concentration values.

Global Equivalence Ratio	Axial Position			Reynolds Number	% maximum variation
	100 mm	200 mm	500 mm		$\frac{Máx_{500} - Máx_{100}}{Máx_{100}}$
0,7	2-30 ppm	4-32 ppm	17-36 ppm	5400	20
1,0	4-36 ppm	5-43 ppm	12-44 ppm	5400	22
1,3	4-38 ppm	3-45 ppm	15-49 ppm	5400	29
0,7	2-11 ppm	2-13 ppm	12-25 ppm	7300	127
1,0	1-16 ppm	2-24 ppm	18-44 ppm	7300	175
1,3	1-22 ppm	2-31 ppm	24-53 ppm	7300	141
0,7	2-12 ppm	2-5 ppm	6-18 ppm	9500	50
1,0	1-7 ppm	2-10 ppm	12-27 ppm	9500	285
1,3	1-10 ppm	3-16 ppm	15-36 ppm	9500	260

### 3.3 Temperature

There are four NO<sub>x</sub> formation mechanisms in a combustion reaction. The thermal mechanism, or Zel'dovich, dominant in temperatures above 1800 K; the prompt mechanism, or Fenimore, associated to the chemistry of hydrocarbons and been particularly important in rich mixture regions; the N<sub>2</sub>O mechanism and the NO formation mechanism through the fuel mechanism. (Turns, 1995). The first two mechanisms are considered the main responsible for the formation of NO during combustion of hydrocarbons without pre-mixing with air (Turns, 1995), presenting different degree of importance of different fuels.

The temperature field may show if there is a predominance between the thermal mechanism and the prompt mechanism. Figures 4 to 6 show the radial temperature profiles versus Reynolds number considering the three longitudinal positions.

H = 100 mm  $\Phi = 1,3$

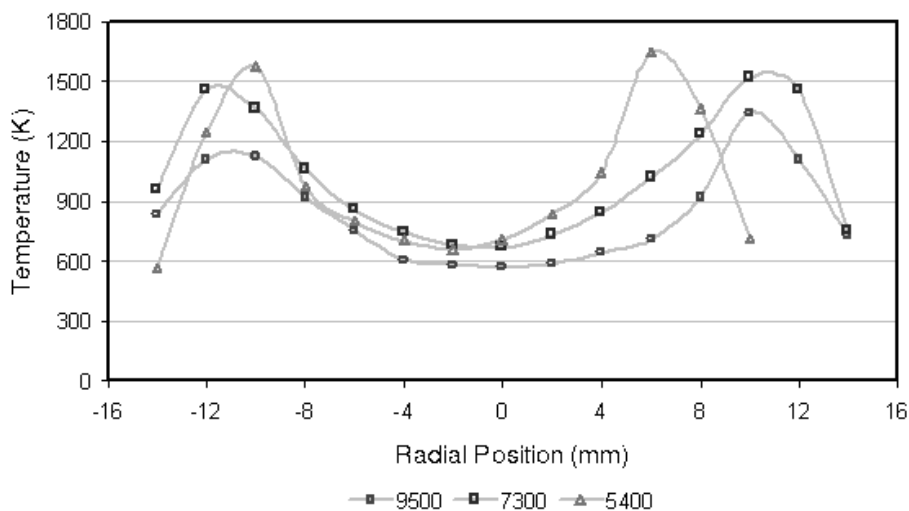


Figure 4. Temperature profile at 100 mm.

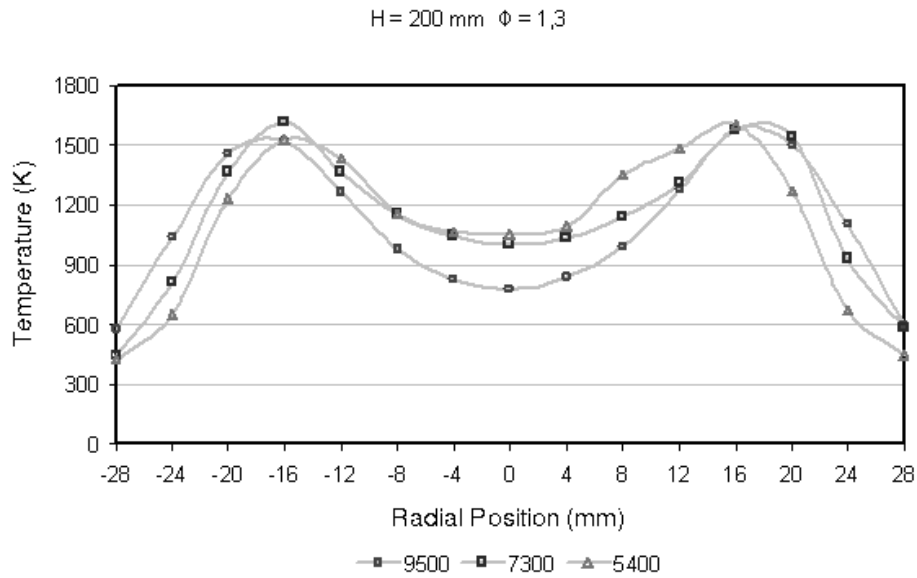


Figure 5. Temperature profile at 200 mm.

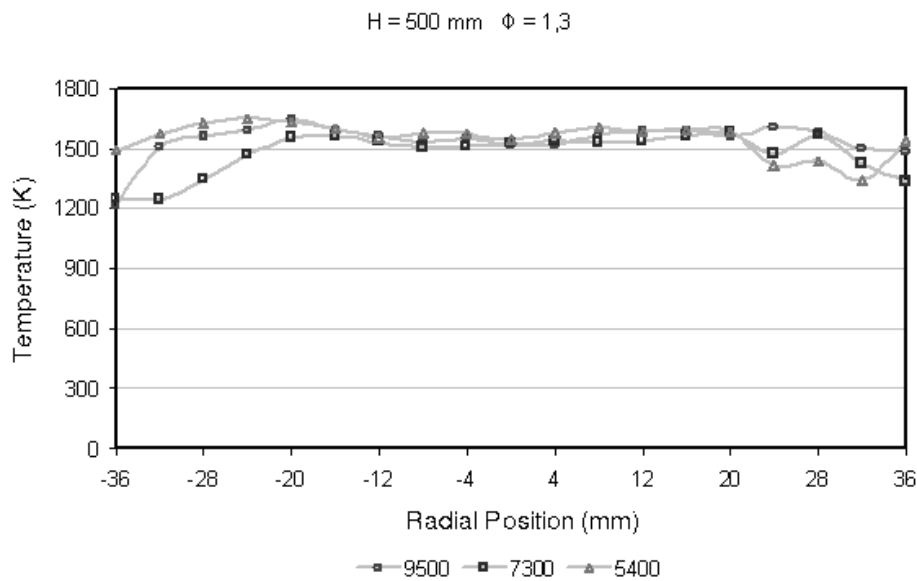


Figure 6. Temperature profile at 500 mm.

The temperature measurements show a predominance of the prompt mechanism over the thermal mechanism, since relatively low average temperatures, never above 1700 K, are found. This tendency is also observed for other global equivalence ratios.

### 3.4 EINO<sub>x</sub> Versus Reynolds Number

A way to quantify the NO<sub>x</sub> emission in non-premixed flames is through the emission index of NO<sub>x</sub> (EINO<sub>x</sub>). The emission index of NO<sub>x</sub> can be expressed in g of NO<sub>x</sub> produced for Kg of fuel burned. Therefore the emission index is given by:

$$EINO_x = \frac{\dot{m}_{NO}}{\dot{m}_{Comb.}} = \frac{\dot{m}_{NO}}{\rho_{Comb.} u_0 \pi d_0^2 / 4} \quad (1)$$

Figures 7 to 9 present the values of EINO<sub>x</sub> [g NO/kg fuel] as a function of the Reynolds number and the tendency lines. The results presented collect data with the same global equivalence ratio and different Reynolds numbers, that is, the fuel jet velocity is increased keeping the same mass ratio air/fuel. The R<sub>2</sub> value, between 0 and 1, reveal the degree of correlation between the estimated values given by the tendency lines and the actual data. The closer R<sub>2</sub> is to unity the greater the reliability of the tendency line.

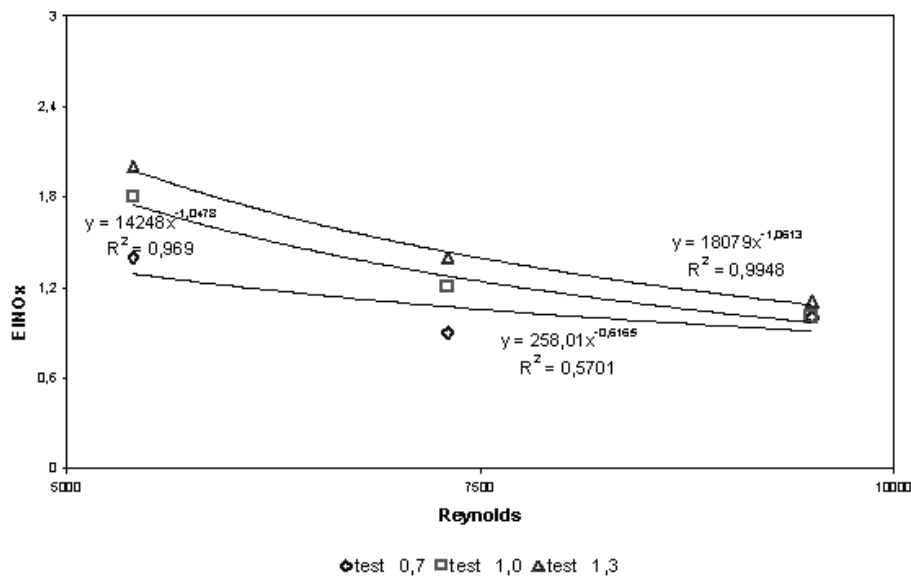


Figure 7. EINO<sub>x</sub> versus Reynolds number, longitudinal position: 100 mm.

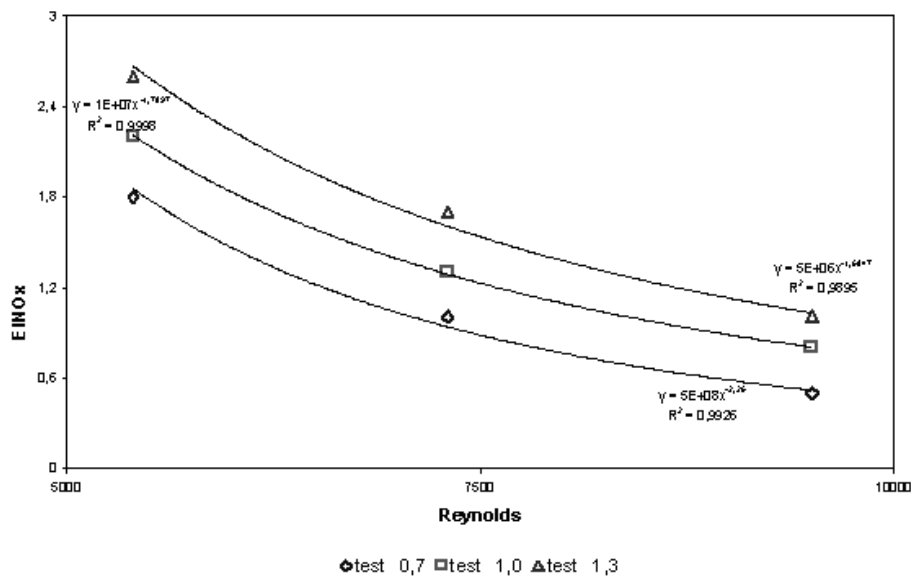


Figure 8. EINO<sub>x</sub> versus Reynolds number, longitudinal position: 200 mm.

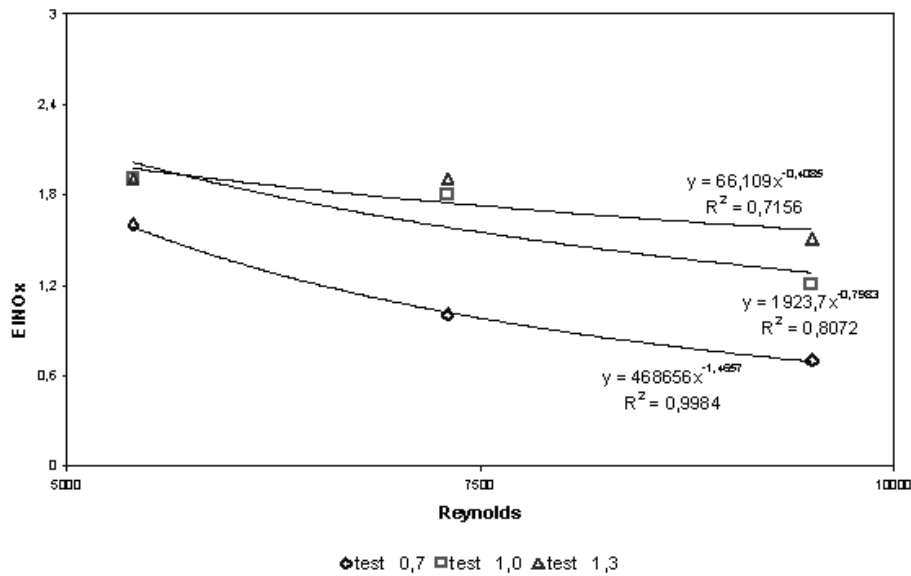


Figure 9. EINOx versus Reynolds number, longitudinal position: 500 mm.

Direct observations from Fig. 7 to 9 show that, for different heights, increasing the Reynolds number the concentration of EINOx is reduced, while increasing the global equivalence ratio increases it. For any global equivalence ratio, in any longitudinal position EINOx concentration is lower with increasing Reynolds number, that is with increasing fuel jet velocity.

#### 4. CONCLUSIONS

Regarding the flame shape the results show that increasing the turbulence levels due to the increase in the jet velocity, the flame thickness is reduced when the surrounding co-axial air jet velocity is increased.

From the temperature measurements it is possible to conclude that there is a predominance of the prompt mechanism over the thermal mechanism, since the average temperature have relatively low values. The temperatures measured were always under 1700 K.

In flames with equivalence ratio of 1.3 the EINOx were higher than the pour and stoichiometric global equivalence ratios for any longitudinal position, confirming results available in the literature.

In any global equivalence ratio and any longitudinal position the EINOx is reduced with the increase in the Reynolds number due to the increase in jet velocity. There are results in the literature that agree with the present finding and others that disagree. The explanation for the results found in the present work may be related to the flame residence time. As the characteristic time is reduced, the time available for the NOx reaction is also reduced.

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#### 6. REFERENCES

- Bilger, R. W. and Beck, R. E. Fifteenth Symposium (Int.) on Combustion, The Combustion Institute, Pittsburgh, p. 541, 1975.
- Caldeira- Pires, A., Carvalho Jr., J. A.; Heitor, M. V. Experimental characterization of nitric oxide formation mechanisms and emission rates in turbulent non-premixed jet flames. Instituto Superior Técnico, Lisboa, Portugal. Personal communication. 1997.
- Caldeira Pires, A.; Heitor, M. V. and Carvalho Jr., J. A. Characteristics of Nitric Oxide Formation Rates in Turbulent Nonpremixed Jet Flames. Combustion and Flame. 120:383-391, 2000.
- De Soete, G.G. Fundamentals of NO Formation and Destruction. Part of the Course Combustion of Solid Fuels, Leeuwehorst Congress Centre, Noordwijkerhout, the Netherlands, September 19-23, 1988.
- De Vries J. E., Study on turbulent fluctuations in diffusion flames. PhD Thesis, Delft University of Technology, 1994.



- Drake, M. C., Correa, S. M., Pitz, R. W., Shyy, W.; Fenimore, C. P. Superequilibrium and thermal nitric oxide formation in turbulent diffusion flames. *Combustion and Flame*, v.69: p. 347-365, 1987.
- Driscoll, J. F., Chen, R. H. and Yoon, Y. Nitric Oxide Levels of Turbulent Jet Diffusion Flames: Effects of Residence Time and Damkohler Number. *Combustion and Flame*, 88: 37-49, 1992.
- Kuo, K. K., Principles of combustion. New York: John Wiley and Sons, 1986.
- Martins, C. A. ; Ferreira, M. A. ; Pimenta, A. P. ; Pires, A. C. ; Carvalho Jr, J. A. . Ch And C2 Radicals characterisation in natural gas turbulent diffusion flames. *Revista Brasileira de Ciências Mecânicas, Brasil*, v. XXVII, p. 110-118, 2005
- Meunier, Ph. Costa, M.; Carvalho, M. G. On NOx emissions from turbulent propane diffusion flames. *Combustion and Flame*, v. 112: p. 221-230, 1998.
- Starner S.H.; Bilger R.W. Characteristics of a piloted diffusion flame designed for study of combustion turbulence interactions. *Combustion and Flame*, v. 61: p.29-38, 1985.
- Santos, A. and Costa, M. Reexamination of the Scaling Laws for NOxB Emissions from Hydrocarbon Turbulent Jet Flames. *Combustion and Flame*. 142: 160-169, 2005.
- Turns, S. R.; Lovett, J. A. Measurements of oxides of nitrogen emissions from turbulent propane jet diffusion flame. *Combustion Science and Technology*, v. 66, p. 233-249, 1989.
- Turns, Stephen R. Understanding NOx formation in nonpremixed flames: experiments and modeling. *Prog. Energy Combust. Sci*, v. 21, p. 361-385,1995.

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