NO_X ABATEMENT FROM OIL COMBUSTION – REBURNING AND LOW NOX BURNER

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Abstract. *The performance of a low NO_x burner and a reburning system for heavy oil combustion has been investigated experimentally. Reburning and burnout simulations were carried out initially in order to determine preliminary experimental parameters for maximum NO_x reduction. The kinetic model was later used to simulate the experimental data in the conditions that showed higher NO_x reduction. The main conclusions are: (1) the implementation of reburning yields a reduction of 63-71% (according to stoichiometric ratio in the reburning zone) in the NO_x emission related to the unstaged fuel value, without detriment of combustion quality; (2) the implementation of air staging by low NO_x burner yields a reduction of 48% in the NO_x emission related to the unstaged value, however the particulate material (PM) emission rates tended upwards; (3) the implementation of reburning in a low NO_x flame yields a reduction of 80% in the NO_x emission related to a base line flame (unstaged fuel and air), however the PM emission rates tended upwards and (4) an unique kinetic model is unable to make a good simulation of reburning and burnout zones even if the injection systems provides good mixing between the primary gas stream, the natural gas and the supplementary air, showing the necessity of a mixing-kinetic model.*

Keywords. *Low NO_x burner, reburning system, heavy oil combustion, reburning and burnout simulations, kinetic model*

1. Introduction

Fuel oil is an important industrial fuel in Brazil, as it participated as 10.6% of the energy consumption in industries in agreement with the 2001 national energy balance. Brazilian fuel oils are produced by Petrobras (Brazilian Petroleum Company) from RASF (asphaltic residue) and RESVAC (vacuum residue) diluted with light fractions at different proportions and having different specifications which differ basically in viscosity and sulfur content. These fuel oils are classified as types 1 to 9 according to their increasing viscosity and as A or B according to their sulfur content, A for the high (above 1%) and B for low one (up to 1%). They have abnormally high viscosity (the type 1 is more viscous than United States residual oil, referred to as Bunker C) and nitrogen content (usually around 1%) and in the burning they tend to produce flue gases with high content of particulate material (PM) and nitrogen oxides (NO_X).

The performance of a low NO_x burner and a reburning system for heavy oil combustion, both designed and built by IPT, was investigated in a horizontal test furnace. Natural gas was used as reburning fuel. Furthermore, a kinetic model was built in order to estimate the initial experimental parameters to higher NO reduction and to obtain a better description of reburning and burnout zones for the trials carried out. This paper reports the activities achieved and the results obtained.

2. Experimental set-up

2.1. Test furnace

The experimental work was carried out in a cylindrical horizontal test furnace of 1.1 m internal diameter and 4.0 m length. It has an internal refractory lining that allows internal surface temperatures to be maintained around 900 ^oC. The furnace steel shell is double plated and divided in nine sections. Each section, as well as the furnace front end are provided with independent water cooling flows. This allows the computation of the net heat flux distribution on the furnace internal surface and the fraction of the total power transferred to it. The combustion chamber is provided with a longitudinal opening where measurement probes can be inserted.

The furnace is coupled to a horizontal channel. This channel is attached to a air preheater, which is coupled to a stack provided with a platform where the sampling station for gas composition and PM collecting is located. In all trials the furnace internal pressure was kept at 25 Pa (effective) to avoid air incoming through leakages.

2.2. Burner

A burner was designed, according to the scaling criteria (constant residence time approach combined with geometric and thermal similarities), and built by IPT to be coupled to the test furnace. The burner can operate either as a conventional one or as a low NOx burner - Fig. (1). The Fig. (2) shows the atomization performance of Y-Jet nozzle attached to it, as examined in the IPT Sprays Laboratory. The main parameters of the burner are:

- nominal power release: 1.4 MW (120 kg/h oil)
- primary air with fixed swirl (S = 0.8) and tertiary air without swirl
- secondary air with variable swirl (S = 0.4 − 3.7)
- primary, secondary and tertiary air with independent registers
- oil gun equipped with atomizer of the type Y-Jet (six outlet holes of 1.3 mm diameter and 30° angle)

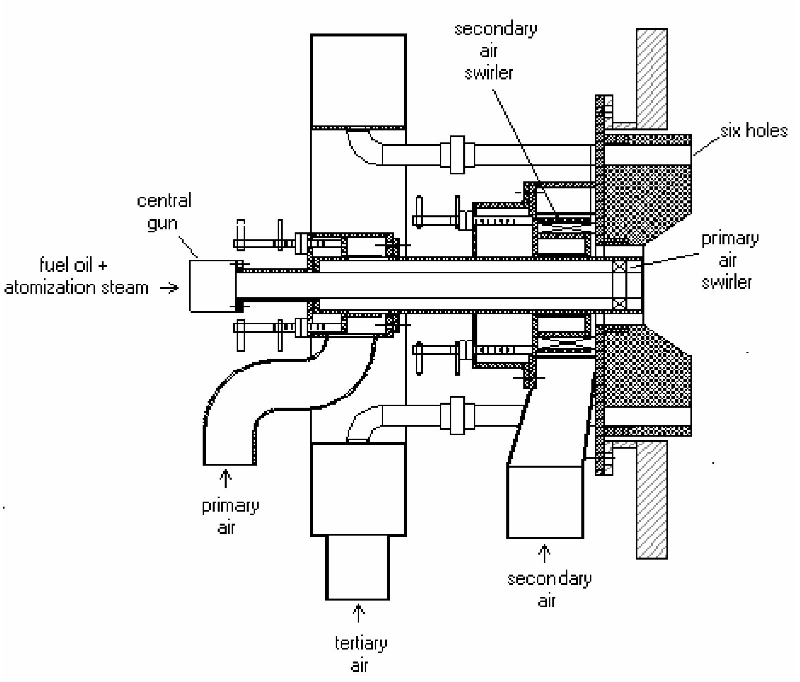


Figure 1. Burner schematic drawing

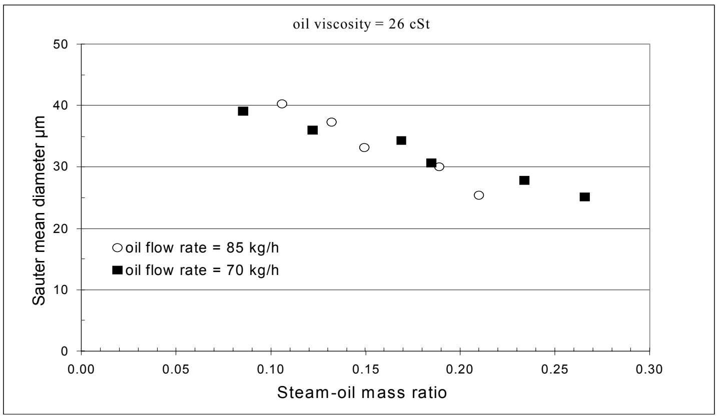


Figure 2. Droplet diameter versus atomization steam-oil ratio

2.3. Reburning system

A reburning system was designed and built by IPT to be installed inside the test furnace. The natural gas and reburning air injectors were designed in order to provide a good mixing of the issuing flows with the primary gas stream, so that three separate sequential zones are created along the furnace (primary, reburning and burnout), and reburning/burnout stoichiometries and residence times are reasonably uniform. This goal was specially difficult to achieve for the injection and mixing of gas due to the low momentum of the jets and the large difference in flow rates with respect to the primary stream.

The natural gas flow was split into 24 radial jets, 30° inclined in relation to the injection section (forward) and was uniformly distributed over the periphery of an annular injector (900 mm internal diameter) coaxial with the combustion chamber and set up at 2.8 m forward nozzle burner. The number, diameters and inclination of jets and injection velocity are selected from a series of cold previous tests. Since the air reburning flow rate is higher, its distribution into the primary flow is much easier than for the reburning fuel. The air flow was split into 12 radial jets, 20^{o} inclined (forward), distributed over an annular injector set up at 3.8 m forward nozzle burner.

In order to provide a better homogenization of the primary stream in the reburning zone inlet a wall of holey refractory bricks was placed in the furnace just before natural gas injection section.

2.4. Instruments

A data acquisition program was provided for monitoring and recording the main operational parameters during the tests. Flow rates, pressures and temperatures were measured with typical instruments - orifice plates, pressure transducers and thermocouples. A suction pyrometer, built by IPT, was used to monitor the gas phase temperature inside the furnace.

Water-cooled probes were inserted into the stack and inside the furnace for gas sampling. The gas flux was continuously pumped across a cooling and filtering system and further sent to a continuous gas analysis train.

The continuous gas analysis set was composed by O_2 (paramagnetic), CO, CO_2 (infrared), VOC - volatile organic compounds (flame ionization), NO_x (chemiluminescence) analysers. The uncertainty of these measurements is ± 3 %, but not less than 5 ppm.

For particulate sampling an isokinetic sampler, built according to the USEPA method n^{o} 5, was used. For sampling procedure the same method was followed. The sizes and morphologies of particles were analyzed through scanning electron microscopy (SEM).

2.5. Fuels

The fuels burned in tests were fuel oil (main fuel) and natural gas (reburning fuel), which properties are shown in Tab. (1).

Table 1. Fuel oil and natural gas characteristics

Fuel oil		**Natural gas**	
Type (Brazilian nomenclature)	4A	Composition (% volume; dry):	
Composition (% mass; dry):		_ methane	89.51
_ carbon	87.6	_ ethane	5.99
_ hydrogen	9.5	_ propane	1.66
_ nitrogen	0.82	_ butane	0.57
_ sulfur	1.1	_ pentane	0.21
_ ash	0.05	_ hexane and heavier	0.09
_ asphaltenes	6.4	_ nitrogen	1.10
_ carbon residue (Conradson)	13.4	_ carbon dioxide	0.88
High heat value (MJ/kg)	42.1	Heat value (MJ/Nm^3):	
Kinematics viscosity (cSt):		_ high	39.8
_ at 80 ^{o}C	1453	_ low	36.0
_ at 150 ^{o}C	43		
Density (g/cm^3):			
_ at 25 ^{o}C	1.03		
_ at 50 ^{o}C	1.02		

3. Tests

3.1. Methodology

In order to investigate the performance of the low NOx burner and the reburning system four groups of trials were carried out. The former group, called "white tests" was carried out burning oil without the application of any NO_x abatement technique (reburning or air staging).

In the second group of trials, called "reburning tests" air and natural gas were supplied to the proper zone of the furnace. During the third group, called "low NOx burner tests", the burner was operated as a low NO_x (injection of primary, secondary and tertiary air). Finally, in the fourth group of trials the reburning and low NOx burner techniques were applied simultaneously.

3.2. White Tests

3.2.1.Results

The effect of air excess over NO_x and CO emissions were investigated - Fig. (3). From these experiments, an O_2 content in flue gases around 2.0% vol.dry was established for these and further tests (primary zone combustion for the reburning tests).

The main white tests results are shown in Tab. (2). The SEM images of the particulate material collected in flue gases can be seen in Fig. (4).

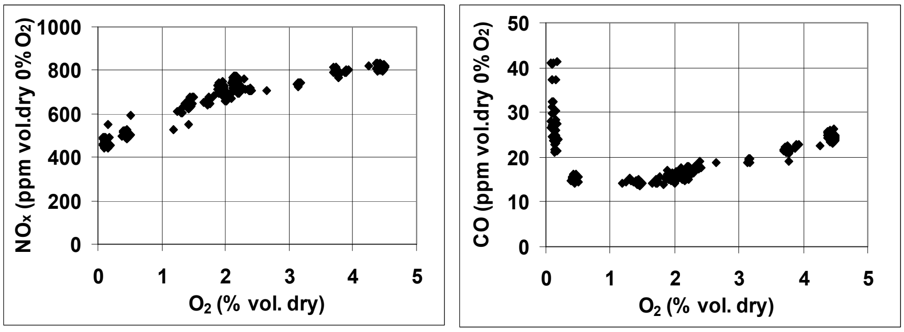


Figure 3. NO_x and CO emissions

Table 2. Characterization of the white tests

Flow rates (kg/h)		**Flue gases composition (dry)**	
Fuel oil	85.0	O_2 (%vol.) ***	2.10
Primary air	308	CO_2 (%vol.)	14.4
Secondary air *	985	CO (ppmvol.0%O_2)	12.2
Atomization steam	10.9	NO_x (ppmvol.0%O_2)	693
Temperatures (°C)		VOC (ppmvol.0%O_2)	0
Fuel oil (burner inlet) **	175	PM (mg/$Nm^3$0%O_2)	42
Air (burner inlet)	285	**Mass ratios**	
Flue gases (stack)	261	1^{ary} air / (1^{ary} + 2^{ary} air)	0.24
		Atomization steam / oil	0.13

* swirl number = 0.4 ** viscosity = 25 cSt *** air coefficient (λ) = 1.11

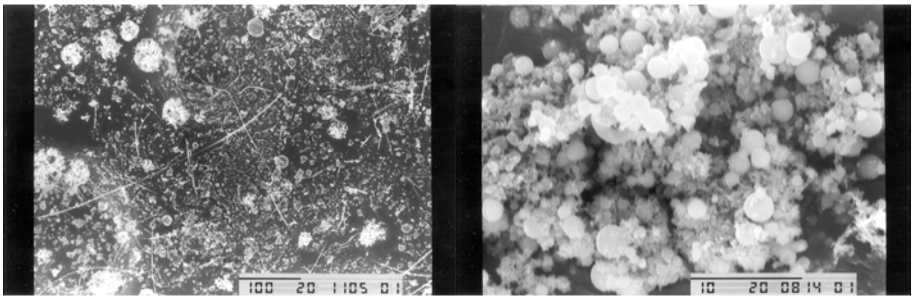


Figure 4. Scanning electron microscopy of particulate material
a) bar length = 100 μm (200 times) b) bar length = 10 μm (3500 times)

3.2.2. Comments

- The burning of a Brazilian typical fuel oil led to an elevated NO_x concentration in the flue gases - 400-800 ppm vol.dry 0%O_2, according to air excess, thus above the established emission standards. There is not a Brazilian standard for NO_x - the USA standard is 277 ppm vol.dry 0%O_2. Thus, the employ of a NO_x abatement technique is advisable.

- In Fig. (3), it can be seen that the NO_x emissions increase with the O_2 concentration in the flue gases and for that values of O_2 content above 0.5% the CO emissions change slightly with the air excess. Since the CO content is directly related to combustion quality, it should be fixed a O_2 in gases of 0.5% vol.dry for these and other tests, but instead 2.0% vol.dry because this value is typical for the operation of large boilers in Brazil. The curves for CO and NO_x versus O_2 agreed very well with those obtained during field measurements carried out by IPT in large boilers firing fuel oils.

- For this O_2 content in flue gases, it is possible to observe that the particulate material emissions are relatively low, down the Brazilian emission standards (124 mg/Nm^3 0%O_2) and CO emissions too (maximum of 53 ppm vol. dry 0%O_2 for a well operated boiler, according USEPA – United States Environmental Protection Agency). This denotes the good combustion quality obtained in tests.

- The observations made using electron microscope - two micrographies are presented in Fig. (4) have shown predominance of relatively small particles (0.6-1.5 μm), spherical, compact and with smoothed surface ("bead" cenospheres) in the flue gases. This allows identifying the collected particulate basically as coke and agrees with the expected, except by particle size (the expectation was by larger particles, in the same diameter range of the oil droplets).

3.3. Reburning Tests

3.3.1. Results

In order to analyses the effect of air coefficient in the reburning zone (λ_R), groups of trials with different values for this parameter was carried out. Due to an experimental error, a group of trials were carried out with an O_2 content in the reburning zone inlet very low (1.3 rather than 2.0%) that led to a λ_R very low too (0.32 rather than around 0.6 intended). The results are shown in Tab. (3). The SEM images of the particulate material collected in flue gases referring to trials group with $\lambda_R = 0.76$ are presented in Fig. (5).

The spatial distribution of main species concentration and gas temperature have been determined in order to allow further understanding of the reburn process and to provide a complete set of data for the validation of kinetic model built. It is shown, in Fig. (6), only the concentrations of NO_x and O_2 for the group of trials with $\lambda_R = 0.32$. The temperature values (not shown here) were in the range between 1100 °C (around the axis and near gas injection section) and 900 °C (around the walls and near air reburning injection section).

3.3.2. Comments

- The implementation of reburning yielded a significant reduction in the NO_x emission (63 to 71%, according to reburning zone coefficient, related to the unstaged fuel value), without detriment of combustion quality. This is illustrated in Fig. (7) and Fig. (8).

- The NO_x USA standard was attended with the application of reburning technology. The "cost" was the partial substitution of the oil by natural gas. In the tests, the natural gas was responsible for around 13-17% of global power release.

- The PM morphology changed by the reburning technique applied. The particles are nearly shallow spheres such as those typical for oil combustion with higher sizes between 20-50 μm. Since the PM present in the flue gases is derived from the combustion oil in both process (with and without reburning) the same size range was expected.

Table 3. Characterization of the reburning tests

	Group of trials		
	A	**B**	**C**
Flow rates (kg/h)			
Fuel oil	74.2	71.2	69.6
Primary air	266	272	251
Secondary air *	849	813	754
Atomization steam	9.4	9.2	9.3
Reburning air	81	181	147
Natural gas	8.5	10.6	11.5
Flue gases composition (dry)			
O_2 (%vol.)	0.9	2.1	0.3
CO_2 (%vol.)	15.0	13.9	16.0
CO (ppmvol.0%O_2)	11	13	17
NO_x (ppmvol.0%O_2)	254	238	167
VOC (ppmvol.0%O_2)	0	0	0
PM (mg/$Nm^3$0%O_2)	44	48	-

	Group of trials		
	A	**B**	**C**
Temperatures (°C)			
Fuel oil (burner inlet) **	174	175	176
Air (burner and injectors inlet)	311	297	317
Flue gases (stack)	261	269	285
Mass ratios			
1^{ary} air / (1^{ary} + 2^{ary})	0.24	0.25	0.25
Atomization steam / oil	0.13	0.13	0.13
Furnace air coefficients (λ)			
Main zone	1.11	1.12	1.06
Reburning zone	0.76	0.67	0.32
Burnout zone	2.38	3.16	1.13
Global	1.04	1.11	1.01
Natural gas power release (% of global)			
Natural gas	12.6	15.8	17.2

* swirl number = 0.4 ** viscosity = 25 cSt

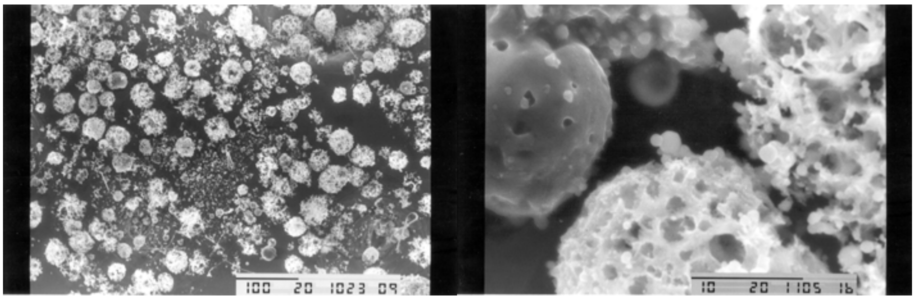
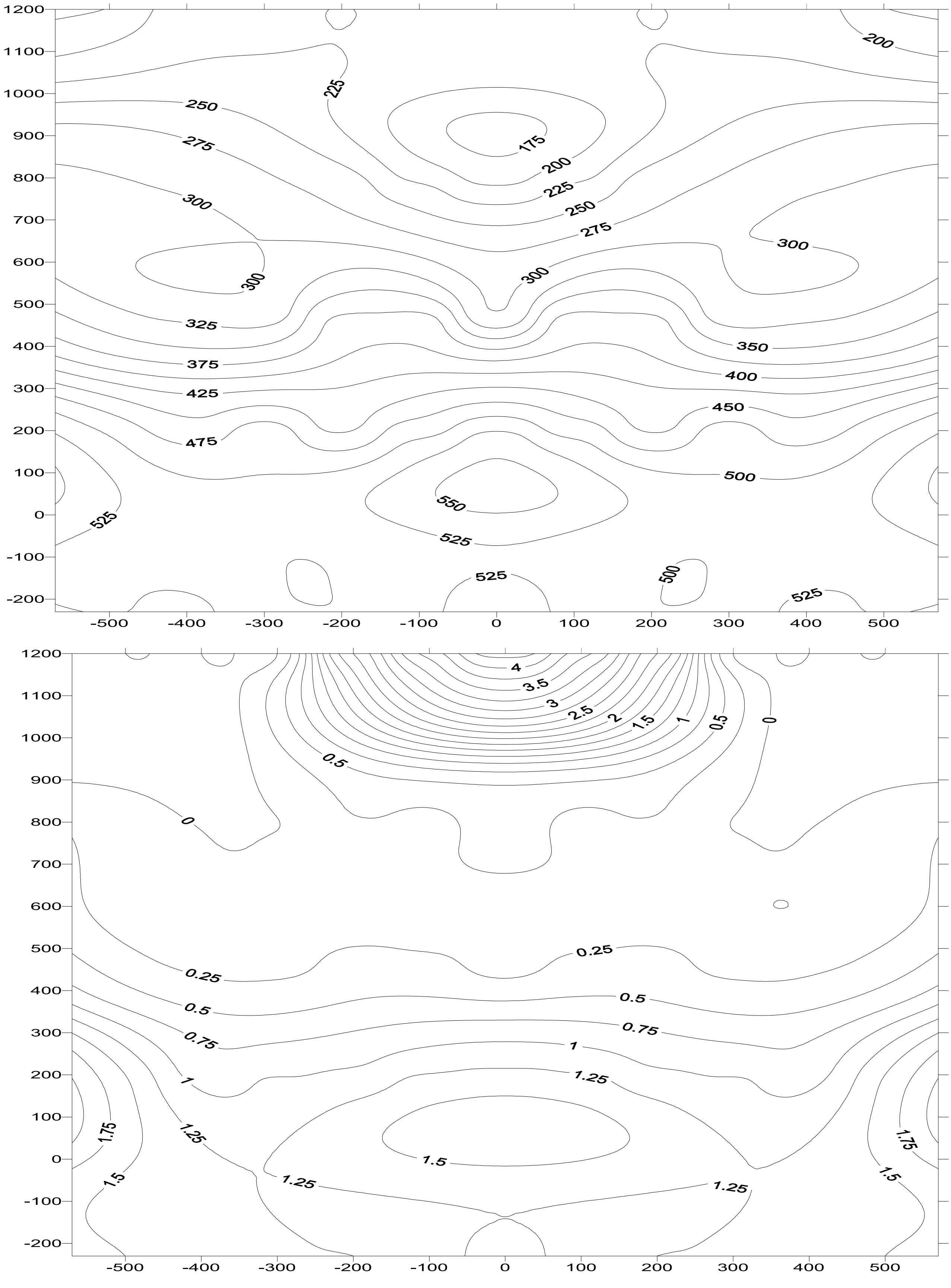


Figure 5. Scanning electron microscopy of particulate material
a) bar length = 100 μm (200 times) b) bar length = 10 μm (3500 times)



axis x: radial distances from centre of furnace (mm)
axis y: axial distances from natural gas injection section (mm)
y = 0 → natural gas injection section   y = 1000 mm → air reburning injection section

Figure 6. Distribution of a) NO_x (ppm) and b) O_2 (%) in the furnace for $\lambda_R = 0.32$

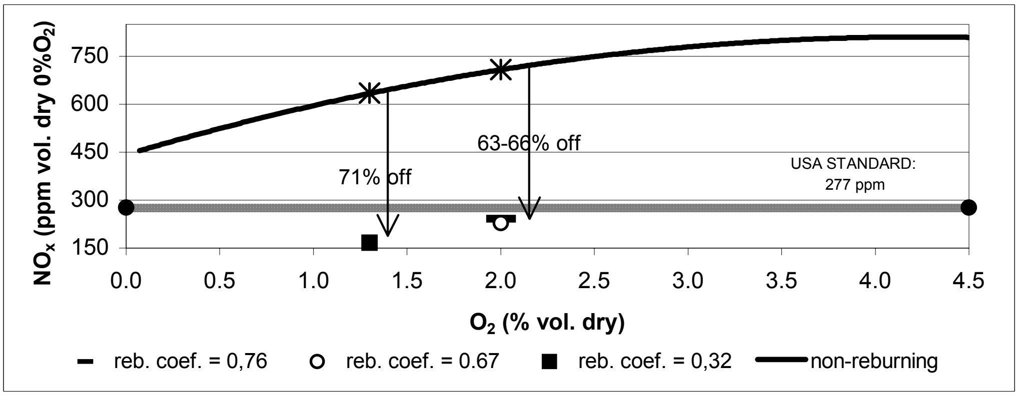


Figure 7. Effect of reburning on NO$_x$ emissions

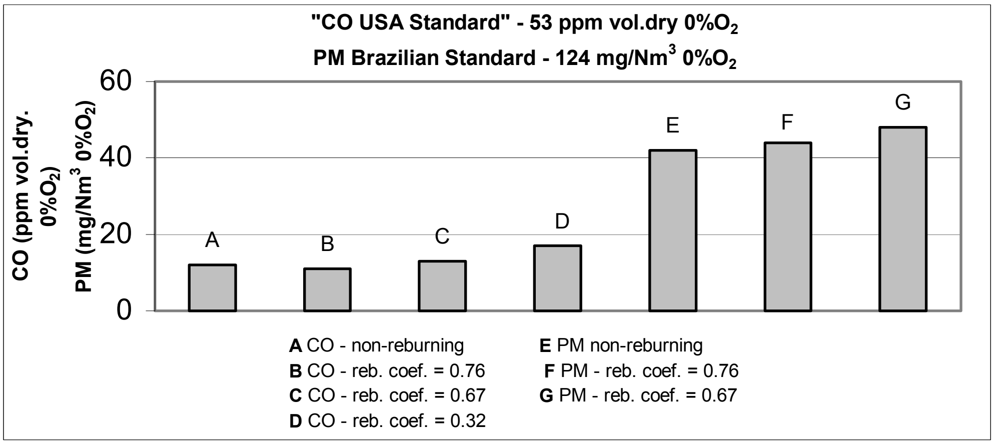


Figure 8. Effect of reburning on CO and PM emissions

- In Fig. (6), it can be seen a fair radial uniformity of O_2 and NO_x concentrations. Unfortunately, for the other conditions (with larger λ_R), the uniformity was worse, probably as a consequence of the lower natural gas flow rates. Thus it is difficult to define a "reburning zone". Nearly, for all conditions, this zone can be defined as the region comprised between the sections of the furnace situated from 200 to 800 mm beyond the natural gas injection. This results in a residence time for the gases in the reburning zone around 550 ms. The definition of a "burnout zone" is still more difficult because the combustion of the remaining fuel took place between the end of the furnace and any point along the flue gases channel.

3.4. Low NOx Burner and Reburning + Low NOx Burner Tests

3.4.1. Results

The effect of (1^{ary} air - total air) and (3^{ary} air - total air) mass ratios in NO$_x$ emissions was investigated - Fig. (9). From these experiments, the following parameters were established:

- 1^{ary} air - total air mass ratio = 0.19
- 2^{ary} air - total air mass ratio = 0.19
- 3^{ary} air - total air mass ratio = 0.62

The main low NOx tests results are shown in Tab. (4).

Reburning + low NOx burner tests were carried out with the burner operated as a low NOx, at the conditions previously established (primary, secondary and tertiary air flow rate of 19, 19 and 62% of the total air, respectively) and with the employment of the reburning technology ($\lambda_R = 0.5$). The main results achieved are shown in Tab. (5).

3.4.2. Comments

- The implementation of low NOx burner yielded a significant reduction in the NO$_x$ emission (48%), although not sufficient to attend the NO$_x$ USA standard. The standard was attended with the simultaneous application of reburning (NO$_x$ reduction of 80%). This is illustrated in Fig. (10).

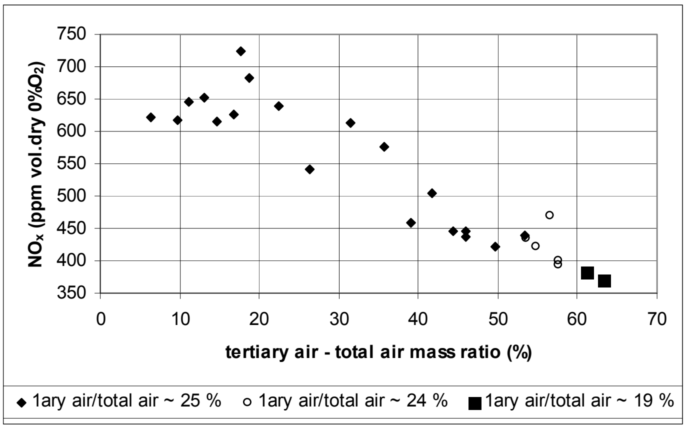


Figure 9. NO_x emissions versus 3^{ary} air - total air ratio

Table 4. Characterization of the low NOx burner tests

Flow rates (kg/h)		**Flue gases composition (dry)**	
Fuel oil	84.9	O_2 (%vol.) ***	2.0
Primary air	228	CO_2 (%vol.)	16.0
Secondary air *	237	CO (ppmvol.0%O_2)	15
Tertiary air	765	NO_x (ppmvol.0%O_2)	359
Atomization steam	10.9	VOC (ppmvol.0%O_2)	0
Temperatures (°C)		PM (mg/Nm^{3}0%O_2)	166
Fuel oil (burner inlet) **	176	**Mass ratios**	
Air (burner inlet)	266	1^{ary} air / (1^{ary} + 2^{ary} + 3^{ary} air)	0.19
Flue gases (stack)	260	2^{ary} air / (1^{ary} + 2^{ary} + 3^{ary} air)	0.19
		3^{ary} air / (1^{ary} + 2^{ary} + 3^{ary} air)	0.62
		Atomization steam / oil	0.13

* swirl number = 0.4 ** viscosity = 25 cSt *** air coefficient (λ) = 1.11

Table 5. Characterization of the reburning + low NOx burner tests

Flow rates (kg/h)		**Mass ratios**	
Fuel oil	71.0	1^{ary} air / (1^{ary} + 2^{ary} + 3^{ary} air)	0.18
Primary air	190	2^{ary} air / (1^{ary} + 2^{ary} + 3^{ary} air)	0.18
Secondary air *	189	3^{ary} air / (1^{ary} + 2^{ary} + 3^{ary} air)	0.64
Tertiary air	671	Atomization steam / oil	0.13
Atomization steam	9.4	**Furnace air coefficients (λ)**	
Reburning air	148	Main zone	1.09
Natural gas **	10.5	Reburning zone	0.49
Temperatures (°C)		Burnout zone	1.68
Fuel oil (burner inlet) ***	178	Global	1.05
Air (burner and injectors inlet)	292		
Flue gases (stack)	262		
Flue gases composition (dry)			
O_2 (%vol.) ****	1.0	NO_x (ppmvol.0%O_2)	139
CO_2 (%vol.)	15.6	VOC (ppmvol.0%O_2)	0
CO (ppmvol.0%O_2)	20	PM (mg/Nm^{3}0%O_2)	224

* swirl number = 0.4 ** natural gas power release (% of global) = 15.7 *** viscosity = 25 cSt **** air coefficient (λ) = 1.10

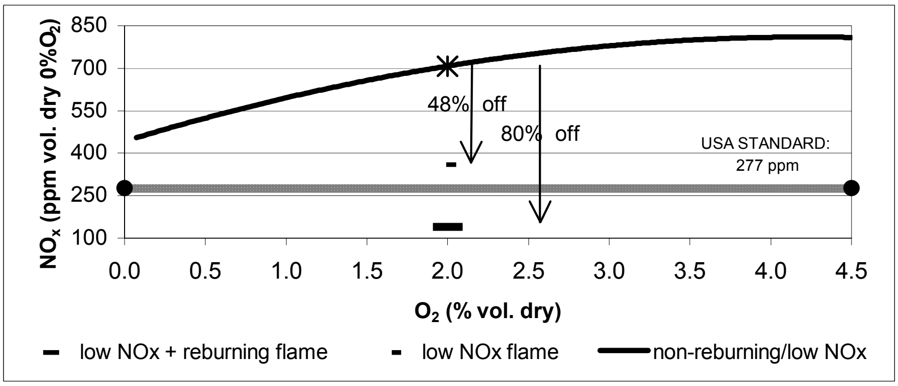


Figure 10. Effect of low NOx burner and low NOx burner + reburning on NO_x emissions

- The implementation of low NOx burner either lonely or together with the reburning led to a sharp increase in the particulate material emissions, far beyond the Brazilian standard. Practically, any change in the CO emissions has not been observed. This is illustrated in Fig. (11).

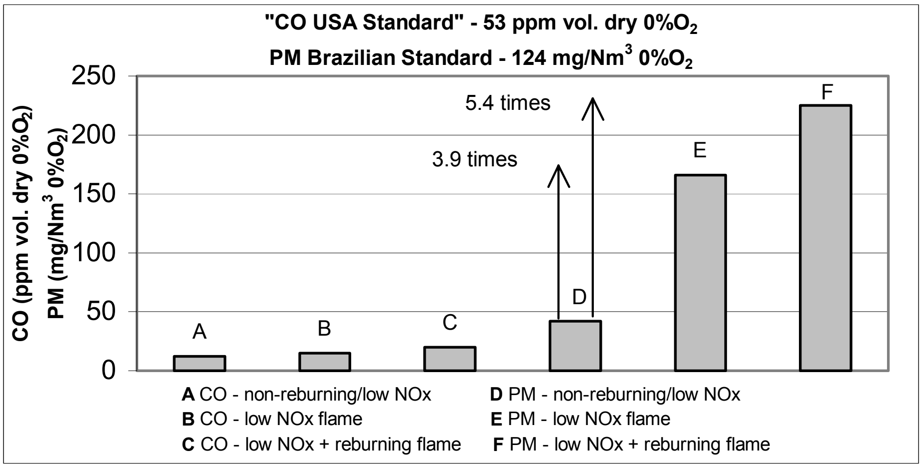


Figure 11. Effect of low NOx burner and low NOx burner + reburning on CO and PM emissions

4. Kinetics simulation

An initial model consisted of 514 reactions and 68 species was previously evaluated with literature data (Braun-Unkhoff, 1999). It was reduced to 347 reactions and 58 species through the production rate and sensitivity analysis and this one was applied to simulate both reburning and burnout kinetics of natural gas for heavy oil combustion. The reaction set was based on Glarborg et all (1998) work and on GRI-3.0 mechanism (Smith). The simulations were made considering methane, ethane (sum of all other hydrocarbon fractions), N_2 and CO_2 fractions as natural gas and using the PLUG code of CHEMKIN-III. Several simulations for both reaction zones established the operational parameters for larger NO_x reduction (Bertran, 2002). The simulated and experimental concentration profiles of NO_x and CO in the reburning zone are shown in Fig. (12).

4.1. Comments

There is not a good agreement with experimental and simulated results for both reburning and burnout zones. The kinetic model is better for the condition with $\lambda_R = 0.32$, in which the temperature is lower and the reburning zone is more homogeneous, making the consumption rate of natural gas more concordant to the experimental. Anyway, the simulation results show that it is necessary to couple a mixing model to the kinetic mechanism to get a better description of reburning and burnout reactions.

5. Conclusions

The main conclusions of the work are:

(1) The implementation of reburning yields a reduction of 63-71% (according to stoichiometric ratio in the reburning zone) in the NO_x emission related to the unstaged fuel value, without detriment of combustion quality.

(2) The implementation of air staging by low NO_x burner yields a reduction of 48% in the NO_x emission related to the unstaged value, however the particulate material (PM) emission rates tended upwards.

(3) The implementation of reburning in a low NO_x flame yields a reduction of 80% in the NO_x emission related to a base line flame (unstaged fuel and air), however the PM emission rates tended upwards.

(4) An unique kinetic model is unable to make a good simulation of reburning and burnout zones even if the injection systems provides good mixing between the primary gas stream, the natural gas and the supplementary air, showing the necessity of a mixing-kinetic model.

6. Acknowledgements

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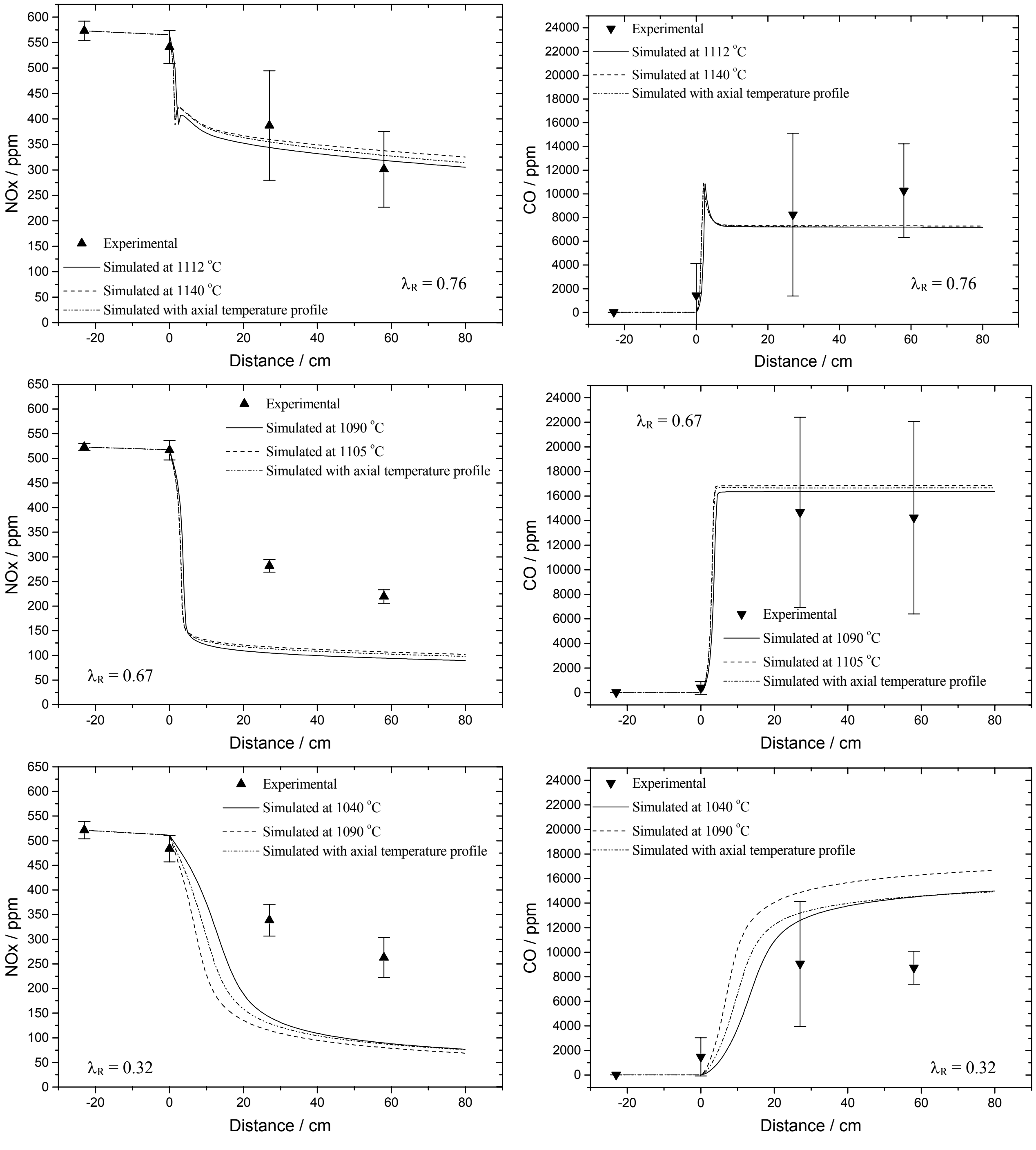

* In the graphs, the distance "0" corresponds to 20 cm after the natural gas injection.

Figure 12. Simulated and experimental concentration profiles of NO_x and CO in the reburning zone

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