# **IMPORTANCE OF THE REBURN FUEL ON THE REDUCTION OF NO**<sub>x</sub> **EMISSIONS FROM A LABORATORY FURNACE**

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**Abstract.** The aim of the present work was to evaluate the effectiveness of the reburning process using biomass (rice husk) as reburn fuel in a large-scale laboratory furnace. For comparisons purposes, tests were also conducted using natural gas and ethylene as reburn fuels. The paper reports new data for flue-gas emissions for a wide range of experimental conditions which quantify the effects of the reburn fuel fraction (energy basis), residence time in the reburning zone and initial NO<sub>x</sub> concentration for the three secondary fuels on NO<sub>x</sub> reduction from the present combustor. The results show that at reburn zone residence times of about 0.7 s the reburning performance of the rice husk: i) is comparable to that of the natural gas reburning at high reburn fuel fractions (low reburn zone stoichiometric ratios), with almost 60% NO<sub>x</sub> reduction achievable at reburn fuel fractions of 25% and 30%, and ii) approaches those of the natural gas and ethylene at high initial NO<sub>x</sub> concentrations between around 500 and 970 ppm. The results also reveal that there is a correlation between the extent of NO<sub>x</sub> reduction and particle burnout: the higher the reduction the lower the burnout. However, the present results combined with previous data from the same furnace using coal as reburn fuel suggest that the use of biomass as reburn fuel in full-scale equipment would have a lower impact in the carbon burnout as compared with coal.

Keywords. NO, reburning, rice husk, natural gas, ethylene.

## 1. Introduction

The formation of nitrogen oxides  $(NO_x)$  is an important pollutant source in combustion systems, being its control still a major issue. Various control strategies are nowadays available to reduce  $NO_x$  emissions from practical combustion systems. Among these techniques, fuel reburning has proved to be an effective way to reduce  $NO_x$  emissions from different combustion systems. The reburning technology involves the establishment of three distinct reaction zones along the height of a furnace. In the primary reaction zone, most of the fuel is burned under normal fuellean conditions. Subsequently, further downstream, in the secondary reaction zone, the so-called reburn zone, the remaining fuel is introduced to establish a fuel-rich reburning zone wherein hydrocarbon radicals (CH<sub>i</sub>) generated from the reburn fuel assist the reduction of part of the  $NO_x$  formed in the primary zone to molecular nitrogen and other nitrogenous species. Finally, still further downstream, in the tertiary reaction zone, overfire air is added to establish overall fuel-lean conditions and, thereby, allow for the burnout of the remaining fuel fragments. A rather complete review on reburning technologies has been provided by Smoot et al. (1998).

Gaseous fuels have been identified as the most effective reburn fuels due to the null or minimal content of nitrogen in their composition and due to their ability to produce easily active hydrocarbon radicals which react with NO (Bilbao et al., 1997). Examination of the literature reveals that most of the work on reburning has been carried out in bench and large-scale combustors using a variety of fossil fuels, chiefly pulverized coals, as reburn fuels, see, for example, Smart and Morgan (1994), Kicherer et al. (1994), Liu et al. (1997), Morgan et al. (1998), Maly et al. (1999), Zhong et al. (2002), Hardy and Kordylewski (2002), Hampartsoumian et al. (2003) and Dimitriou et al. (2003), among others. In general, these studies revealed that  $NO_x$  reductions up to about 60-70% may be accomplished with about 20-25% of the total heat input coming from the reburn fuel provided the NO<sub>x</sub> concentrations from the primary reaction zone are relatively high (typically above 500 ppm). In most cases, however, reburning using coal as reburn fuel might have a serious impact in the carbon burnout over a wide range of furnace operating conditions. Under these circumstances, the use of biomasses as reburn fuels may be a good alternative to coals without a deleterious effect on particle burnout performance due to its higher content of volatile matter as compared with coal, which incidentally favors NO<sub>x</sub> reduction in the case of the use of coal as reburn fuel, see, for example, Smart and Morgan (1994), Kicherer et al. (1994) and Morgan et al. (1998), among others. Furthermore, from an environmental viewpoint, biomass fuels present a number of advantages compared with conventional fossil fuels. Apart from its renewable nature, biomass fuels present lower levels of sulphur and, usually, toxic metals, and are CO<sub>2</sub> neutral. Moreover, the reburn fuel usually represents only around 20-25% of the total heat input so that relatively small quantities of biomass are required. Despite all these facts, little attention appears to have been devoted to methodical investigations to  $NO_x$  control through biomass reburning. The few exceptions available in the open literature include the studies of Maly et al. (1999) and of Harding and Adams (2000). The latter authors have conducted a rather comprehensive study using two types of wood, whose reburning effectiveness was compared with those of natural gas and coal. Overall, their experimental results suggested that woods are as effective as natural gas or coal as a reburning fuel.

The aim of the present work was to evaluate the effectiveness of the reburning process using biomass (rice husk) as reburn fuel in a large-scale laboratory furnace. For comparisons purposes, tests were also conducted using natural gas and ethylene as reburn fuels. The paper reports new data for flue-gas emissions as a function of the reburn fuel type; for each reburn fuel its  $NO_x$  reduction potential was analyzed as a function of the reburn fuel fraction (energy basis), residence time in the reburning zone and  $NO_x$  concentration at the exit of the primary zone (initial  $NO_x$  concentration).

#### 2. Test facility and methods

The tests were carried out in the Instituto Superior Técnico (IST) large-scale laboratory furnace. A major advantage of the present furnace is that it is large enough to ensure that the essential physics of full-scale combustors are simulated. In particular, it is large enough to ensure fully turbulent flow combined with significant thermal radiation transfer, but small enough to enable the collection of detailed and reliable data.

The combustion chamber is cylindrical in shape. Its axis is vertical to minimize asymmetry due to natural convection and biased ash particle deposition and it is down-fired to facilitate particulate removal. The cylinder comprises eight water-cooled steel segments each 0.3 m in height and 0.6 m in internal diameter. The roof section and the upper four segments are lined with a layer of refractory, and a ceramic fiber blanket sandwiched between the refractory and the water-cooled jacket. Each segment incorporates a pair of diametrically opposed 0.22-m-diameter ports for probing and injection of reburn fuels and overfire air. A more detailed description of the experimental facility can be found elsewhere (Casaca and Costa, 2003).

Figure (1) shows schematically the arrangement of the top end wall of the furnace and of the reburn system. The burner geometry is typical of that used in power stations for wall-fired boilers and consists of a burner gun and a secondary air supply in a conventional double-concentric configuration, terminating in an interchangeable refractory quarl of half-angle 30°. In this work, the burner gun used consisted of two concentric tubes: the central orifice was used for the introduction of primary air and the annular orifice was used for the introduction of natural gas and ammonia (see section 4). The secondary air entered a plenum chamber situated above the burner, in which it encountered a moveable block swirl generator of the type developed at the International Flame Research Foundation. The air then flowed through an interchangeable cylindrical duct and, subsequently, into the refractory quarl section. The primary air was supplied by a fan of variable speed and its flow rate was measured using a calibrated orifice plate installed upstream of the fan. The natural gas was supplied by the Portuguese gas company via the IST grid and the ammonia from standard bottles. In both cases, the gas flow was controlled with pressure regulators and valves and the flow rates measured using calibrated rotameters.

The reburn system consisted of two independent, but similar, devices: the reburn fuel injection system and the overfire air injection system. The secondary fuels (rice husk, natural gas and ethylene) and the overfire air were injected radially inside the combustion chamber at the furnace axis through similar nozzles, as shown in Fig. (1). Other nozzle configurations were tested for the introduction of the reburn fuels and overfire air into the combustion chamber, namely, angled radial injection along the furnace centerline. They showed, however, poorer performances as compared with the present injection configuration, which enabled, on the one hand, an even distribution and adequate penetration of the reburn fuels into the primary zone products, without particles impact on the furnace walls, and, on the other hand, ensured that, in the burnout zone, the overfire air was well mixed with the gaseous products exiting the reburn zone.

In an initial set of experiments, the locations of all reburn fuels and overfire air injection were varied along the furnace centerline and the flue-gas concentrations measured. The outcome of this procedure in conjunction with detailed in-flame measurements of  $O_2$ , CO,  $CO_2$ , unburnt hydrocarbons (HC) and  $NO_x$  concentrations, and of gas temperatures without reburning (Casaca and Costa, 2003) allowed us to define appropriate locations for the injection of all reburn fuels (see section 4).

The biomass was transported to the reburn fuel injection system via a loss-in-weight feeder and a compressed air ejector system. Trials using nitrogen, instead of air, as the reburn fuel transport medium were also carried out. The solid fuels feeding system is fully described elsewhere (Casaca and Costa, 2003). The transport air for biomass was supplied by an air compressor (10 bar) and the overfire air by a fan of variable speed with both flow rates being measured using calibrated rotameters. The nitrogen and ethylene were supplied from standard bottles with the gas flow controlled with pressure regulators and valves and the flow rates measured using calibrated rotameters.

## 3. Experimental techniques and uncertainties

The gases for the measurement of the flue-gas data ( $O_2$ , CO, CO<sub>2</sub>, HC and NO<sub>x</sub> concentrations) were withdrawn using a water-cooled stainless steel probe. The probe was composed of a centrally located 3-mm-i.d. tube, through which samples were evacuated, surrounded by two concentric tubes for probe cooling. The probe outlet was connected to a heated polytetrafluorethylene line through which the gases were conducted first to a cyclone separator immersed in an ice bath, to eliminate the moisture and the larger particulates from the gas sample, followed by a microfiber filter, to remove the remaining particulates, and then on to the analyzers. The analytical instrumentation included a magnetic pressure analyzer for  $O_2$  measurements, nondispersive infrared gas analyzers for  $CO_2$  and CO measurements, a flame ionization detector for HC measurements and a chemiluminescent analyzer for  $NO_x$  measurements. The analog outputs of the analyzers were transmitted via analogic/digital boards to a computer where the signals were processed and the mean values computed. Zero and span calibrations with standard mixtures were performed before and after each measurement session. The maximum drift in the calibration was within  $\pm 2\%$  of the full-scale.

At the furnace exit, where the gas composition was nearly uniform, probe effects were likely to be negligible and errors arose mainly from quenching of chemical reactions and sample handling. Samples were quenched near the probe tip to about 150 °C and condensation of water within the probe was avoided by controlling the inlet temperature of the cooling water (typically to around 60 °C). Repeatability of the flue-gas data was, on average, within 5%.



Figure 1. Furnace roof, burner arrangement and reburn system.

Solids sampling was performed with the aid of a stainless steel water-cooled, water-quenched probe, details of which may be found in Casaca and Costa (2003). Upon leaving the probe, the char samples were collected in a glass container. Following decantation, the collected wet solid sample was placed in an oven at approximately 110 °C to dehydrate. Complete dehydration was ascertained by repeated drying and weighing of the sample until the measured mass became constant. The solid samples were subsequently analyzed (ultimate analysis). The particle burnout data were obtained from the following equation:

$$\Psi = \frac{1 - \left(w_k / w_x\right)}{1 - w_k} \tag{1}$$

where  $\Psi$  is the particle burnout, w is the dry ash mass fraction, and the subscripts k and x refer to ash content in the input solid fuel and char sample, respectively.

Uncertainties in particle burnout calculations based on the use of ash as a tracer are connected to ash volatility at high heating rates and temperatures and ash solubility in water. In order to determine the accuracy of the experimental procedure and measurements, carbon and oxygen elemental mass balances have been performed for all tested conditions. The overall mass balance discrepancies were always below 3%. Radial traverses at the exit sampling location indicated no spatial variation in particle burnout and repeatability was, on average, within 10%.

## 4. Experimental conditions

In this work, natural gas was used as the primary fuel for all trials; reburn fuels included biomass (rice husk), natural gas and ethylene. The main characteristics of the rice husk and of the hydrocarbon gaseous fuels are given in Table 1, which also includes a summary of the furnace operating conditions for the reburning trials. They encompass various experimental flames which quantify the effects of the reburn fuel fraction (energy basis), residence time in the reburning zone and initial  $NO_x$  concentration for the three secondary fuels on  $NO_x$  reduction from the present combustor.

| (a) <u>Characteristics of rice husk</u>                  |               |                          |              |                         |           |
|--|---------------|--------------------------|--------------|-------------------------|-----------|
| Proximate analysis (wt %)                                |               | Ultimate analysis (wt %) |              | Particle size (% under) |           |
| Moisture   | 9.8           | Carbon                   | 38.8         | 30 µm                   | 4.0       |
| Volatiles  | 59.9          | Hydrogen                 | 4.6          | 70 µm                   | 11.6      |
| Fixed Carbon   | 14.7          | Nitrogen                 | 1.3          | 100 µm                  | 17.6      |
| Ash  | 15.6          | Sulphur                  | 0.3          | 300 µm                  | 54.2      |
|  | (             | Oxygen                   | 29.6         | 500 µm                  | 77.9      |
| High Heating Value (MJ/kg) 15.0                          |               | 5.09                     |              | 1000 µm                 | 97.6      |
| Low Heating Value (MJ/kg) 13.88                          |               | 3.88                     |              | 2000 µm                 | 100       |
| (b) Characteristics of hydrocarbon gaseous fuels         |               |                          |              |                         |           |
| Composition (vol. %)                                     |               | Natural Gas              |              | Ethylene                |           |
| $ CH_4$  |               | 83.7                     |              | -                       |           |
| $C_2H_5$   |               | -                        |              | 99.5                    |           |
| $C_2H_6$   |               | 7.6                      |              | -                       |           |
| $\geq C_3$   |               | 2.9                      |              | -                       |           |
| $N_2$  |               | 5.4                      |              | 0.5                     |           |
| $CO_2$   |               | 0.2                      |              | -                       |           |
| He   |               | 0.2                      |              | -                       |           |
| High Heating Value (MJ/kg)                               |               | 5                        | 53.8         | 50.3                    |           |
| Low Heating Value (MJ/kg)                                |               | 4                        | 48.6 47.2    |                         |           |
| (c) <u>Furnace operati</u>                               | ng conditions |                          |              |                         |           |
| Parameter  |               |                          | Reburn Fuels |                         |           |
|  |               |                          | Natural Gas  | Ethylene                | Rice Husk |
| Total thermal input (kW)                                 |               |                          | 130-200      | 130-200                 | 130-186   |
| Primary zone stoichiometric ratio                        |               |                          | 1.1          | 1.1                     | 1.1       |
| Reburn zone stoichiometric ratio                         |               |                          | 0.72-1.1     | 0.7-1.1                 | 0.75-1.1  |
| Burnout zone stoichiometric ratio                        |               |                          | 1.1          | 1.1                     | 1.1       |
| Reburn fuel fraction (%)                                 |               |                          | 0-35         | 0-35                    | 0-30      |
| Reburn zone residence time (s)                           |               |                          | 0.31-0.92    | 0.31-0.93               | 0.32-0.92 |
| NO <sub>2</sub> concentration at primary zone exit (ppm) |               |                          | 50-970       | 50-970                  | 50-970    |

Table 1. Characteristics of the fuels and furnace operating conditions for the reburning tests

Throughout the experiments, the conditions of the main flame were kept constant, in particular, the secondary air swirl number of the primary flame was maintained at 1 and the stoichiometric ratio in the primary zone (from burner exit to reburn fuel injection) at 1.1. The stoichiometric ratio in the burnout zone (from overfire air injection to furnace exit) was also kept constant (also equal to 1.1) for all experiments through the regulation of the flow rate of the overfire air.

The stoichiometric ratio of the reburn zone (from reburn fuel injection to overfire air injection), defined as the total air-fuel ratio divided by the stoichiometric air-fuel ratio of all air and fuel in the primary and reburning zones, was varied through the control of the reburn fuel mass flow rate, whose value is used to define the reburn fuel fraction, *i.e.*, the fraction of reburn fuel injected as a percentage of the total thermal input.

The residence time in the reburn zone was varied by altering the location of the injection of the overfire air (for biomass:  $0.59 \ m \le Z_2 \le 1.15 \ m$ ; for hydrocarbon gaseous fuels:  $0.61 \ m \le Z_2 \le 1.17 \ m - Z_2$  is defined in Fig. 1) with respect to the location of the reburn fuel injection (for biomass:  $Z_1 = 0.33 \ m$ ; for hydrocarbon gaseous fuels:  $Z_1 = 0.37 \ m - Z_1$  is defined in Fig. 1). Note that when biomass was used as the reburn fuel, transport air was required, in contrast with its needless in the case of the hydrocarbon gaseous fuels. The presence of oxygen in the transport fluid increases the reburn stoichiometry which may adversely affect the NO<sub>x</sub> reduction. In order to evaluate the impact on reburning performance of the presence of oxygen in the carrier fluid tests have also been carried out using molecular nitrogen as the transport medium. The results of these trials are presented and discussed in the following section.

The initial NO<sub>x</sub> levels, *i.e.*, NO<sub>x</sub> concentrations at the exit of the primary zone, were set by doping the natural gas fed to the burner with small amounts of ammonia to simulate fuel-NO formation in addition to the prompt- and thermal-NO produced in the natural gas flame. Values of  $Z_1 \ge 0.33 m$  ensured high burnout of the primary fuel and complete conversion of the doped NH<sub>3</sub> to NO within the primary zone. This is a critical goal in reburning technology not only to ensure that NO<sub>x</sub> generated from the main fuel is completed in the primary zone but also to prevent fractions of volatile-N (or char-N in the case of solid fuels) to be transported into the reburn combustion zone.

# 5. Results and discussion

Before examining the data, some considerations concerning the NO reduction mechanisms that occur under reburning conditions are required in order to better understand the discussion of the results presented below. Usually, NO reduction during reburning occurs via its reactions with CH<sub>i</sub> and NH<sub>i</sub> radicals. The partial oxidation and pyrolisis of the hydrocarbon reburn fuel results in the formation of CH<sub>i</sub> radicals that react with NO as follows (Myerson, 1975):

$$NO + CH_i \rightarrow HCN + \dots$$
(2)

Hydrogen cyanide produced via reaction (2) is converted to  $N_2$  through the following sequence of reactions (Thorne et al., 1986):

$$HCN + O \rightarrow NCO + H$$

$$NCO + H \rightarrow NH + CO$$
(3)
(4)

$$NH + H \rightarrow N + H_2 \tag{5}$$

$$N + NO \rightarrow N_2 + O \tag{6}$$

The sequence of reactions described above is favored under the fuel-rich conditions encountered in the reburning zone. In fact, under fuel-lean conditions, hydrocarbon radicals react with oxygen and/or hydroxyl radicals to form CO via the following reaction:

$$CH_i + O \rightarrow CO + H + \dots \tag{7}$$

Consequently, reactants in reactions (2) and (7) consume  $CH_i$  competitively. A goal in reburning optimization is to maximize the exposure of NO to  $CH_i$  and minimize  $CH_i$  interaction with oxygen (Smoot et al., 1998).

Additional reactions in the reburning zone may result in the formation of NH and NH<sub>2</sub>, which can further reduce NO concentrations through the following reaction (Wendt et al., 1973):

$$NO + NH_i \rightarrow N_2 + H_2O \tag{8}$$

Note that if high concentrations of HCN and NH<sub>3</sub> formed in the reburning zone enter the fuel-lean burnout zone they might be oxidized to NO with the effectiveness of the reburning process suffering accordingly. This situation is likely to arise under high reburn fuel fractions and/or low reburn zone residence times.

As mentioned earlier, in order to evaluate the influence of the carrier fluid on the reburning effectiveness, trials using both air and nitrogen as the reburn biomass fuel transport media were carried out. Fig. (2) shows the results from these trials as a function of the reburn fuel fraction for the biomass studied. In each of the cases presented in the figure, the residence time in the reburning zone was 0.7 s and the initial NO<sub>x</sub> level was 350 ppm.

Figure (2) reveals that at low reburn fuel fractions (below 20%) the reburning effectiveness is higher when the reburn transport medium is nitrogen. At high reburn fuel fractions (25% and 30%) the reburning performance is,

however, independent of the transport medium. This suggests that at low reburn fuel fractions the use of air as the transport fluid yields oxygen concentrations in the reburn zone that largely exceed those required to convert HCN to  $N_2$ , via reactions (3) to (6), so that the reburning mechanism is ineffectual. On the contrary, at high reburn fuel fractions the reducing atmosphere necessary for effective reburning exists, regardless of the reburn fuel transport medium. The present results are consistent with those reported by Morgan et al. (1998) using coal as reburn fuel but not entirely consistent with those reported by Harding and Adams (2000) using wood as reburn fuel. The latter authors found no noticeable effect on NO<sub>x</sub> reduction of the amount of oxygen in the carrier flow, regardless of the reburn fuel fraction. In the present study, the use of  $N_2$ , instead of air, as the transport fluid has, however a negative impact on particle burnout, as will be demonstrated below. In light of this and of the relatively small differences in reburning performance observed at high reburn fuel fractions, the data reported below to evaluate the effects of the residence time in the reburning zone and initial NO<sub>x</sub> concentration on NO<sub>x</sub> reduction were obtained using air as the reburn fuel transport medium at a constant reburn fuel fraction of 20%.



Figure 2. Effect of the reburn fuel transport fluid on the NO<sub>x</sub> reduction for biomass reburning as a function of the reburn fuel fraction. Initial NO<sub>x</sub> concentration: 350 ppm. Reburn zone residence time: 0.7 s.

The reburn fuel fraction, combined with the stoichiometric ratio of the primary zone (kept constant throughout the present experiments) defines the stoichiometric ratio of the reburn zone. In spite of being slightly dependent on the composition of the reburn fuel, the stoichiometric ratio of the reburn zone is around 1 for a value of the reburn fuel fraction of 10%, around 0.9 for 20% and around 0.8 for 30%. Fig. (3) shows the effect of the reburn fuel fraction (or reburn zone stoichiometric ratio) on the NO<sub>x</sub> reduction for the three reburn fuels studied. In each of the cases presented in the figure, the residence time in the reburning zone was 0.7 s and the initial NO<sub>x</sub> level was 350 ppm.

With respect to the effect of the reburn fuel type, Fig. (3) reveals that the impact of ethylene on  $NO_x$  reduction exceeds that of the natural gas in about 10%, regardless of reburn fuel fraction, which is consistent with the findings of Bilbao et al. (1997), who evaluated the reburning performance of various hydrocarbon fuels in a drop-tube reactor. These authors concluded that the effectiveness for  $NO_x$  reduction follows the sequence: acetylene, ethylene, ethane, natural gas and methane that correspond to the hydrocarbon oxidation sequence.

The figure also shows that the reburning performance of the biomass is inferior to that of the hydrocarbon gaseous fuels, regardless of reburn fuel fraction. In contrast with the present results, Maly et al. (1999) have reported data wherein the performance of a biomass (fir lumber wood waste) was superior to that of a natural gas. Apart from the differences in the furnace geometries, natural gas compositions (marginal) and biomass particle sizes distributions (also marginal), this can be mainly attributed to the higher value of their biomass volatile matter content (75.29 wt%) and lower nitrogen content (0.53 wt%), as compared with the present biomass (see Table 1), revealing the direct impact on NO<sub>x</sub> reduction of those two parameters. These observations are also applicable when comparing our data with that of Harding and Adams (2000), referred to in section 1, although in this case direct assessment is difficult owing to important differences in furnace configurations (U-furnace in their case).

With respect to the effect of the reburn fuel fraction, Fig. (3) discloses that the reburning performance of the biomass is comparable to that of the natural gas reburning at high reburn fuel fractions (low reburn zone stoichiometric ratios), with almost 60% NO<sub>x</sub> reduction achievable at reburn fuel fractions of 25% and 30%. At these high reburn fuel fractions of biomass, sufficient hydrocarbon radicals and N-species intermediates are available in the reburn zone to attain NO<sub>x</sub> reductions, via reactions (2) and (8), comparable to those reached with natural gas reburning. Note that in the

case of reburn fuels with nitrogen in their composition, like the present biomass, there will be a critical upper limit of the reburn fuel fraction beyond which the reburning effectiveness shall drop because significant residual fuel-nitrogen will be able to escape from the reburn zone into the burnout zone for subsequent oxidation to NO by the overfire air.

At low reburn fuel fractions (below 20%), the reburning performance of the biomass drops significantly simply because the amounts of biomass used as reburn fuel are insufficient to establish the crucial fuel-rich reburning zone and thereby to generate active hydrocarbon radicals for the reburn mechanism to be effective. As mentioned earlier, under these conditions (fuel-lean) reaction (7) is strongly favored.

The effects of the reburn fuel fraction on reburning performance observed in Fig. (3) are generally in agreement with previous findings using biomass as the reburn fuel, see Maly et al. (1999) and Harding and Adams (2000).



Figure 3. Effect of the reburn fuel fraction on the NO<sub>x</sub> reduction for the three reburning fuels. Initial NO<sub>x</sub> concentration: 350 ppm. Reburn zone residence time: 0.7 s.

Figure (4) shows the effect of the reburn zone residence time on the NO<sub>x</sub> reduction for the three reburning fuels studied. In each of the cases presented in the figure, the reburn fuel fraction was 20% and the initial NO<sub>x</sub> level was 350 ppm. As can be noticed, residence times in the reburning zone from 0.31 s to 0.93 s were studied, on the one hand, because previous investigations (Chen et al., 1986) have indicated that residence times of at least 0.3 s are required for reburning to be effective and, on the other hand, because typical reburn zone residence times in full-scale equipment, limited by boiler access and burnout requirements, are in the range of 0.4-0.7 s.

In regard to the effect of the reburn fuel type, it is seen in Fig. (4) that, with reference to natural gas, ethylene performs better and biomass performs worse throughout the range of reburn zone residence time studied, which is consistent with earlier observations based on Fig. (3).

Figure (4) also shows that for all reburn fuels the percentage of  $NO_x$  reduction increases with the increase of the reburn zone residence time, which is in agreement with previous findings using biomass as the reburn fuel, see Maly et al. (1999) and Harding and Adams (2000). This increased  $NO_x$  reduction with the residence time is simply due to the longer exposure times between the hydrocarbon radicals and the  $NO_x$ . Note that, in general, the differences between the biomass and the hydrocarbon gaseous fuels performances are larger at shorter residence times, say below 0.4-0.5 s, because the former needs time for devolatilization to occur. As a result, at low residence times, oxidation of higher amounts of residual fuel nitrogen will take place in the burnout zone with the reburning performance suffering accordingly.

Figure (5) shows the effect of the initial  $NO_x$  concentration on the  $NO_x$  reduction for the three reburning fuels tested. In each of the cases presented in the figure, the reburn fuel fraction was 20% and the residence time in the reburning zone was 0.7 s. With respect to the effect of the reburn fuel type, it is interesting to note in Fig. (4) that, at high initial  $NO_x$  concentrations, the reburning performance of the biomass approaches that of the hydrocarbon gaseous fuels, with nearly 60%  $NO_x$  reduction attainable at initial  $NO_x$  concentrations between around 500 and 970 ppm. However, at low initial  $NO_x$  concentrations, below around 400 ppm, with reference to natural gas, ethylene performs better and biomass performs worse.

In regard to the effect of the initial  $NO_x$  concentration on the  $NO_x$  reduction, Fig. (5) shows that for initial  $NO_x$  concentrations below around 350 ppm the  $NO_x$  reduction percentage declines, and for  $NO_x$  concentrations above around

500 ppm variations in performance are insignificant, which is in close agreement with the previous findings using biomass as the reburn fuel reported by Maly et al. (1999).



Figure 4. Effect of the reburn zone residence time on the NO<sub>x</sub> reduction for the three reburning fuels. Initial NO<sub>x</sub> concentration: 350 ppm. Reburn fuel fraction: 20%.



Figure 5. Effect of the initial  $NO_x$  concentration on the  $NO_x$  reduction for the three reburning fuels. Reburn fuel fraction: 20%. Reburn zone residence time: 0.7 s.

Figure 6 shows the relation between  $NO_x$  reduction and particle burnout for a number of experimental conditions in which biomass was used as the reburn fuel. For comparison purposes the figure also includes data obtained using nitrogen as the reburn fuel transport medium, revealing the negative impact on particle burnout of the use of  $N_2$ , instead of air, as the biomass transport fluid.

As can be seen in Fig. (6) there is a correlation between the extent of  $NO_x$  reduction and particle burnout: the higher the reduction the lower the burnout. It is important, however, to recall that only the roof section and the upper four segments of the furnace are lined with a layer of refractory (see section 2). Under the present experimental conditions, the heterogeneous reaction zones of the flames are extended well into the water-cooled lower end of the furnace with particle burnout suffering accordingly. In any case, comparable reburning tests carried out in the present furnace using a relatively high volatile coal as reburn fuel yielded particle burnout below 80%. In light of the present results it can be anticipated that reburning using biomass as reburn fuel in full-scale equipment would have a lower impact in the carbon burnout as compared with coal.



Figure 6. Relation between NO<sub>x</sub> reduction and biomass particle burnout.

Finally, it should be stressed that CO and HC emissions were monitored for all experimental conditions reported above. CO emissions were found to be always below 35 ppm and HC emissions always below 7 ppm, regardless the reburn fuel. No relationship was observed between CO and reburn fuel type, reburn fuel fraction, reburn zone residence time, initial  $NO_x$  concentration or flue-gas  $NO_x$  emission.

# 6. Conclusions

The aim of the present work was to evaluate the effectiveness of the reburning process using biomass (rice husk) as reburn fuel in a large-scale laboratory furnace. For comparisons purposes, tests were also conducted using natural gas and ethylene as reburn fuels. The paper reports new data for flue-gas emissions for a wide range of experimental conditions which quantify the effects of the reburn fuel fraction (energy basis), residence time in the reburning zone and initial  $NO_x$  concentration for the three secondary fuels on  $NO_x$  reduction from the present combustor. The main conclusions drawn from this study include:

- 1. At reburn zone residence times around 0.7 s:
  - a) the reburning performance of the rice husk is comparable to that of the natural gas reburning at high reburn fuel fractions (low reburn zone stoichiometric ratios), with almost 60% NO<sub>x</sub> reduction achievable at reburn fuel fractions of 25% and 30%.
  - b) the reburning performance of the rice husk approaches those of the natural gas and ethylene at high initial  $NO_x$  concentrations, with nearly 60%  $NO_x$  reduction attainable at initial  $NO_x$  concentrations between around 500 and 970 ppm

2. There is a correlation between the extent of  $NO_x$  reduction and particle burnout: the higher the reduction the lower the burnout. However, the present results combined with previous data from the same furnace using coal as reburn fuel suggest that the use of biomass as reburn fuel in full-scale equipment would have a lower impact in the carbon burnout as compared with coal.

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