## NUMERICAL STUDY OF HYDROGEN MILD COMBUSTION

by

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In this article a combustor burning hydrogen and air in mild regime is numerically studied by means of computational fluid dynamic simulations.

All the numerical results show a good agreement with experimental data.

It is seen that the flow configuration is characterized by strong exhaust gas recirculation with high air preheating temperature. As a consequence, the reaction zone is found to be characteristically broad and the temperature and concentrations fields are sufficiently homogeneous and uniform, leading to a strong abatement of nitric oxide emissions.

It is also observed that the reduction of thermal gradients is achieved mainly through the extension of combustion in the whole volume of the combustion chamber, so that a flame front no longer exists ("flameless oxidation").

The effect of preheating, further dilution provided by inner recirculation and of radiation model for the present hydrogen/air mild burner are analyzed.

Key words: hydrogen, flameless combustion, computational fluid dynamics

### Introduction

During the past decade and especially in these very last years the abatement of emissions of pollutants has been representing a relevant topic of research. Special emphasis has been particularly given to those novel combustion regimes which have the intrinsic potential to lessen pollution.

Mild oxidation is a promising combustion technique for the reduction of pollutant emissions, which is achieved by lowering the temperature increase in the process [1-7].

Its main principle of operation lies on massive exhaust gas recirculation, so that the temperature increase of reactants and the oxygen concentration can be strongly reduced [8-13].

It has been held that preheating of combustion air causes significant increase in nitric oxide emissions, due to high temperatures [1, 2, 5, 9, 13]. However, in circumstances of low oxygen concentration it is possible to observe a substantial suppression of pollutant emissions [1, 2, 5, 8, 11].

Nevertheless, in presence of such high dilution levels, combustion can take place in stable way only as long as the mixture is preheated higher than the auto-ignition temperature [1, 5, 7, 14].

Hence, it is possible to introduce the following definition for the mild combustion regime [1]: "A combustion process is named mild when the inlet temperature of the reactant mixture is higher than mixture auto-ignition temperature whereas the maximum allowable temperature increase with respect to inlet temperature during combustion is lower than mixture auto-ignition temperature".

The gap existing between the apparently contrasting goals of minimization of pollutant emissions and improvement of process efficiency (regenerative and recuperative burners) can be potentially lowered in mild combustion because of high levels of dilution characteristic of the mixture (*i. e.*, low oxygen concentrations) leading to a strong reduction of temperature peaks in the combustion chamber.

In mild regime reactions occur at lower rates (higher characteristic combustion times) than in traditional processes, so that the characteristic times of chemical kinetics and fluid-dynamics are almost of the same order of magnitude and the Damkohler number is very close to unity [10, 15].

The combustion zone is no longer concentrated close to the flame front but is extended over the whole combustion chamber, resulting in complete disappearance of any luminous emission: hence the name "flameless combustion" [3, 4, 12, 13, 16]. Moreover, it is noticed that this volumetric distributed combustion approaches the mixing conditions of a well stirred reactor [1, 7, 10, 12, 15, 16].

## Combustion burner and numerical model

In the present paper Derudi's [8, 10, 11, 17] laboratory-scale burner for mild combustion has been numerically reproduced in the case of pure hydrogen as a fuel. The experimental apparatus, shown in fig. 1, is basically a close quartz cylinder divided in two sections, a lower one, attaining the air preheating zone (fig. 1, D) and an upper one, where the combustion chamber is located (fig. 1, A).

Both sections are enclosed with refractory insulation (fig. 1, H, G) equipped with electrical resistances.

The burner is characterized by high internal recirculation achieved by a proper aerodynamic design of the combustion chamber. As described in detail by Cavigiolo [8] and Galbiati [11] the air flux vitiated with nitrogen enters the preheating zone (fig. 1, E, D), heats up and flows through the inlet of a high velocity nozzle 80 mm long (fig. 1, B).

The fuel is injected perpendicularly (fig. 1, C) into the nozzle via a capillary pipe (1 mm internal diameter). Before the reactants reach the combustion chamber a partial premix of fuel and air streams occurs in the nozzle but no reaction takes place because of the short residence time.

A secondary inlet of air (fig. 1, F) is also provided and used during the startup of the system because, before switching to flameless conditions, the combustion chamber has to be preheated using conventional flame stabilized on the nozzle tip. The top of the combustor has a small central hole and three larger ones eccentrically located, so that the combustor has a rotational symmetry of 120°.

Temperature measurements were obtained by three thermocouples inserted through these holes and

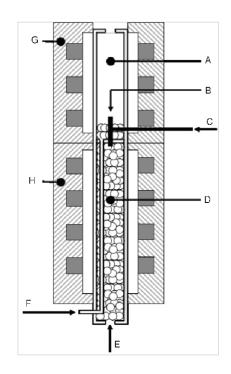


Figure 1. Sketch of the laboratory-scale mild burner used by Derudi *et al.* [8, 10, 11, 17]

located at distance of 100, 170, and 300 mm above the nozzle outlet (the first one was 10 mm shifted from the centreline). NO concentration at the burner exit was carried out via a gas analyzer (water was condensed before entering the analyzer using a water cooled heat exchanger).

Since the numerical simulations of the present paper aim to reproduce a slice of  $120^{\circ}$  of the combustion chamber only it is necessary to calculate the flux properties at the outlet of the nozzle (*i. e.*, entering the combustion chamber). This can be done using a one-dimensional model, where the hypotheses of ideal, steady, adiabatic, non viscous flow are considered:

$$\dot{m}_3 \quad \dot{m}_1 \quad \dot{m}_2 \tag{1}$$

$$\dot{m}_1 u_1 \quad p_1 A_1 \quad \dot{m}_2 u_2 \quad p_2 A_2 \quad \dot{m}_3 u_3 \quad p_3 A_3$$
 (2)

$$\dot{m}_3 c_{P_3} T_3 \quad \dot{m}_1 c_{P_1} T \quad \dot{m}_2 c_{P_2} T_2$$
 (3)

$$\begin{array}{lll} \dot{m}_{3}Y_{3,\mathrm{H}_{2}} & \dot{m}_{2}Y_{2,\mathrm{H}_{2}} \\ \dot{m}_{3}Y_{3,\mathrm{O}_{2}} & \dot{m}_{1}Y_{1,\mathrm{O}_{2}} \\ \dot{m}_{3}Y_{3,\mathrm{N}_{2}} & \dot{m}_{1}Y_{1,\mathrm{N}_{2}} & \dot{m}_{2}Y_{2,\mathrm{N}_{2}} \end{array} \tag{4}$$

$$p_{\rm i} \quad \rho_{\rm i} \, {\rm R} T_{\rm i} \, \frac{Y_{\rm i, H_2}}{W_{\rm H_2}} \, \frac{Y_{\rm i, O_2}}{W_{\rm O_2}} \, \frac{Y_{\rm i, N_2}}{W_{\rm N_2}}$$
 (5)

In eq. (1)-(5)  $\dot{m}_i$  is the mass flow rate,  $p_i$  – the pressure,  $T_i$  – the temperature,  $r_i$  – the density, R – the universal gas constant,  $A_i$  – the area,  $u_i$  – the velocity,  $Y_{i,H_2}$  – the hydrogen mass fraction, and  $W_{H_2}$  – the hydrogen molecular weight. The air flux is represented by i=1, the fuel flux is denoted by i=2 and the mixture entering into the combustion chamber (nozzle outlet) by i=3.

In fig. 2 the temperature at the outlet of the nozzle (*i. e.*, at the combustion chamber inlet) as a function of preheating temperature is shown. The solid line is relative to the analytical model of eqs. (1)-(5). The dots are relative to numerical results of the mixing device obtained with 3-D

simulations, performed by Fluent 6.3, where k- $\varepsilon$  turbulence model [18] was used.

It can be observed a good agreement between the model and the results of the numerical simulations.

The complete homogeneity of the mixture at the injection nozzle's outlet was checked and verified.

Since the preheating oven efficiency  $\eta$  is not precisely known, the error associated with this uncertainty can be estimated by a sensitivity analysis of the burner inlet temperature  $T_3$  with  $\eta$  (where  $\hat{T}_1$   $\eta T_1$  is the preheating oven temperature). It was observed that the maximum error associated with this preliminary calculation is

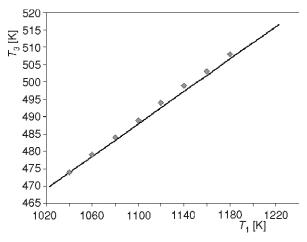


Figure 2. Burner inlet temperature as a function of air preheating temperature (solid line – analytical model, dots – numerical simulations)

Table 1. Preheating temperature, upper oven temperature and internal recycle factor for the five experimental conditions simulated in this work

	$\hat{T}_1$ [°C]	T <sub>wall</sub> [°C]	$K_{ m v}$
1	950	800	25.3
2	950	700	20.3
3	950	700	15.4
4	1100	700	13.0
5	1000	700	9.9

Table 2. Mass flux, temperature,  $O_2$  and  $H_2$  mass fractions of the mixture entering into the combustion chamber ( $N_2$  mass fraction is the complement to 1)

	<i>m</i> [kgs <sup>-1</sup> ]	T[K]	$Y_{\mathrm{O}_2}$	$Y_{ m H_2}$
1	6.44.10-5	517	3.09·10 <sup>-2</sup>	3.77·10 <sup>-3</sup>
2	4.93·10 <sup>-5</sup>	579	4.04·10 <sup>-2</sup>	4.93·10 <sup>-3</sup>
3	3.42·10 <sup>-5</sup>	702	5.82·10 <sup>-2</sup>	7.11·10 <sup>-3</sup>
4	2.65·10 <sup>-5</sup>	903	7.47·10 <sup>-2</sup>	9.12·10 <sup>-3</sup>
5	1.72·10 <sup>-5</sup>	1151	1.16·10 <sup>-1</sup>	1.41·10 <sup>-2</sup>

about 4% for a preheating oven efficiency  $\eta = 0.9-0.95$  [18].

In this work five experimental conditions from Derudi [10] have been simulated. The preheating temperature, the upper oven temperature, and the recirculation factor are reported in tab. 1.

The internal recirculation factor is one of the most important parameters that strongly influences the performance of a mild combustion burner and can be defined as [8, 10, 11, 17]

$$K_V = \frac{(\dot{m}_{\rm air} \quad \dot{m}_{\rm fuel} \quad \dot{m}_{\rm inert})\zeta}{\dot{m}_{\rm air} \quad \dot{m}_{\rm fuel}}$$
(6)

where  $\dot{m}_{\rm air}$ ,  $\dot{m}_{\rm fuel}$ , and  $\dot{m}_{\rm inert}$  are the air mass flow rate, the fuel mass flow rate, and the inert mass flow rate, respectively;  $\zeta = 5$  [8, 10, 11, 17] is the maximum recycle factor due to aerodynamic configuration.

The properties of the mixture entering the combustion chamber (*i. e.*, mass flux, temperature, and composition) resulting from previous calculations with the analytical model are reported in tab. 2.

The numerical simulations were developed modelling a 120° angular section of the burner. *k-\varepsilon* turbulence

model, eddy dissipation concept for chemistry-turbulence interaction [19] and  $P_1$  radiation model [20] have been adopted. From the chemistry point of view, the Warnatz [21] detailed

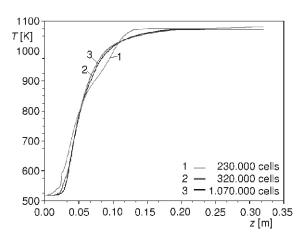


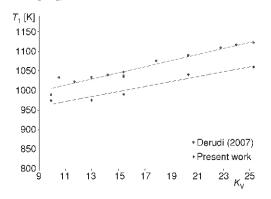
Figure 3. Grid independence analysis: temperature calculated at burner centerline (first case of tab. 1)

mechanism (37 reactions) was used. To take into account Zeldovich [22] thermal NO production and NO formation through NO<sub>2</sub> creation/destruction pathway [24] further 10 reactions was considered.

Before performing all the simulations pertaining the experimental cases, the solution independence from the mesh (which is an unstructured hybrid tetrahedral/hexahedral one) was verified for the first case of tab. 1, using three different meshes: 230.000, 320.000, and 1.070.000 cells (fig. 3). It was found that using the 320.000 cells mesh the solution independence can be considered as sufficiently verified.

#### Results and discussion

Figures 4-7 show the comparison between experimental data and numerical results, in terms of probes' temperatures and of NO emissions. As it can be observed the difference is about 4-5% and therefore it is of the same order of magnitude of the error by which the properties of the mixture entering into the chamber are affected. It is observed that the error due to thermocouples for temperature measurements was about 50 K (Derudi, personal communication [18]).



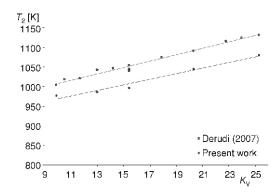
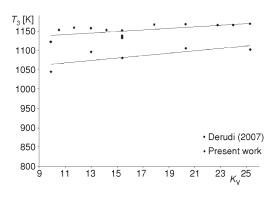


Figure 4. First probe temperature (z = 0.3 m, r = 0)

Figure 5. Second probe temperature (z = 0.17 m, z = 0)

It is worth noting (see fig. 7) the capability of the numerical model to well reproduce the levels of NO emission at the burner exit (few ppmv, for high values of the internal recirculation factor).



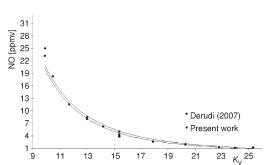


Figure 6. Third probe temperature (z = 0.1 m, r = 0.01 m)

Figure 7. NO emissions [ppmv] at the burner outlet

In fig. 8 streamlines coloured by temperature in a traversal plane inside the combustion chamber are shown, for the first case reported in tab. 1. The picture sheds light on the importance of recirculation (represented by the two main central vortices) by which the reactants are further heated prior to the combustion region. This is mainly achieved by means of heat transfer from burnt products, which are recirculated to mix with the incoming mixture. Moreover it can be no-

ticed from fig. 8 that exhaust gases are entrained by the nozzle which operates as jet pump, so that the incoming stream with its high momentum is diluted by the recirculating burnt products: a new mixture exhaust gases, air, fuel, and inert gases is then formed and heated up continuously.

The effect of further dilution of the mixture due to internally recirculated hot products results in additional preheating of reactants and in lowering oxygen concentration: these two features both affect the flame behaviour thus producing a volumetric combustion. Such a change of the flame structure yields to low Damkohler numbers (around unity), so that quite slow reactions take place.

From observation of figs. 8-10 it is possible to notice a widely distributed reaction region and no more a concentrated combustion as it happens in ordinary combustion processes [5]. In these operative conditions the heat release due to reactions is distributed, yielding to a considerably low temperature rise in the combustion chamber and, as a consequence, to an abrupt suppression of NO emissions.



Figure 8. Streamlines coloured by temperature [K]

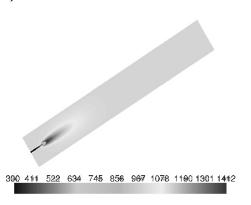


Figure 10. Temperature [K] in a traditional process (first case of tab. 1, with  $T_0 = 300$  K at the inlet)

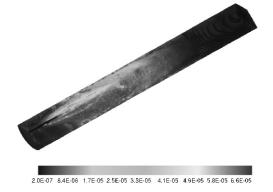


Figure 9. Isosurfaces of OH mass fraction

In traditional processes a well defined flame front, which involves sharp gradients of temperature and reactants concentration, is observed. In mild combustion regime, instead, a volumetric combustion takes place. Such a combustion mode, since it is primarily based on strong dilution levels, can be stably sustained only whenever the reactants are above the self-ignition temperature [1].

If this condition is satisfied then the combustion process results intrinsically stable and does not admit the possibility to switch between to two different combustion states [15]. Ignition and extinction phenomena no longer exist and a monotonic shift unburned to burned condition occurs [7, 12, 16].

It was shown experimentally that, for the hydrogen/oxygen system, in the intermediate temperature range (850-1200 K) a competition between the two main chemical kinetic paths reported in tab. 3 takes place [21, 23, 25, 26]. The second kinetic path prevails in high diluted mixture condition since the oxygen concentration in the first chain branching reaction is reduced

and the efficiency of third body in this path is strongly increased (because of the large amount of burnt gases in the mixture).

Thus in mild combustion regime the second path dominates and fuel self-ignition is due to the destruction of radical which leads to the formation of two radicals [1, 21, 23] through:

Table 3. Hydrogen/oxygen main competing kinetic paths

First kin	etic path	Second kinetic path	
O <sub>2</sub> + H	OH + O	$H + O_2 + M$ $HO_2 + M$	
$H_2 + O$	OH + H	$H_2O_2 + M$ $OH + OH + M$	
H <sub>2</sub> + OH	$H_2O + H$	$H_2 + HO_2 \qquad H_2O_2 + H$	

$$H_2O_2 + M \qquad OH + OH + M \tag{7}$$

Therefore the characteristic time of reaction (7) may be assumed as the representative time for chemistry in hydrogen mild combustion:

$$\tau_{\text{chem}} = \frac{\rho Y_{\text{H}_2\text{O}_2}}{\dot{\omega}_{\text{H}_2\text{O}_2}} \tag{8}$$

Calculating the local fluid dynamic time by:

$$\tau_{\text{fluid}} = \frac{\sqrt[3]{V_{\text{C}}}}{u}$$
 (9)

where  $V_{\rm C}$  is the computational cell volume, it is possible to define the local Damkohler number as:

$$Da = \frac{\tau_{\text{fluid}}}{\tau_{\text{chem}}}$$
 (10)

that is found to be always less than unity in the flow field simulated.

In the present study the effects of recirculation and preheating, and the influence of the numerical model have been assessed. The main contribution for the establishment of hydrogen mild combustion seems to be that of incoming mixture preheating, as shown in fig. 10 (which refers to the first case of tab. 1), as long as lower inlet temperatures are used flameless combustion no longer takes place and a flame front is observed (and the system is characterized by higher temperature peaks).

Recirculation surely plays an important role but does not happen to contribute as much in the present case: from present results and from further simulations with different recirculation factors it is deduced that the inlet mixture is sufficiently diluted and preheated before entering into the combustion chamber. The further preheating and dilution due to internal recycle are found to be not as essential.

As far as the numerical model is concerned it is observed that deactivating the radiation model the solution does not vary appreciably. This is due to the fact that the only absorbing/radiating species is  $H_2O$ , whose absorption coefficient is small. Hence it may be deduced that in hydrogen mild combustion radiant energy transfer is not as important as in hydrocarbon mild combustion.

## **Conclusions**

The present paper deals with main chemical and physical aspects of hydrogen mild combustion. The analysis of such a combustion mode for pure hydrogen as a fuel is led by means

of numerical simulations which reproduce Derudi's experimental observations on a small scale apparatus.

The results are in good agreement with experimental data and illustrate the main features of flameless combustion, showing a substantial absence of any ignition and extinction phenomena.

As a consequence all the reactions are no longer concentrated in the flame front but a volumetric combustion takes place in the burner. High temperature chemical kinetics pathway are in disadvantage with respect to chain terminating reactions: the latter are characterized by local chemical times lower than fluid dynamic times, so that Damkohler number is around one.

The effect of preheating and recirculation has been analyzed carrying out further simulations with different values of such parameters: the main contribute is observed to be due to preheating.

Turning off the radiation model does not considerably affect the numerical solution of the test case.

## **Nomenclature**

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c_{\rm p}
           - specific heat capacity, [Jkg<sup>-1</sup>K<sup>-1</sup>]
                                                                                      W_{\rm H_2} – hydrogen molecular weight, [kgmol<sup>-1</sup>]
           - Damkohler number ( \tau_{\rm fluid} \tau_{\rm chem}^{-1})
                                                                                                 - nitrogen molecular weight, [kgmol<sup>-1</sup>]
           - recirculation factor

    oxygen molecular weight, [kgmol<sup>-1</sup>]

               [ (\dot{m}_{air} \ \dot{m}_{fluel} \ \dot{m}_{inert}) \zeta (\dot{m}_{air} \ \dot{m}_{fuel})^{-1}],
                                                                                      Greek letters
               [-]
\dot{m}_{\rm air}
           - air mass flow rate, [kgs<sup>-1</sup>]
                                                                                                  - preheating oven efficiency, [-]
           - fuel mass flow rate, [kgs<sup>-1</sup>]

 density of mixture i, [kgm<sup>-1</sup>]

                                                                                      \rho_{\rm i}
           - mass flow rate of mixture i, [kgs<sup>-1</sup>]
                                                                                                 - characteristic chemical time, [s]
                                                                                      \tau_{\mathrm{chem}}

inert mass flow rate, [kgs<sup>-1</sup>]
pressure of mixture i, [Pa]

\dot{m}_{\rm inert}
                                                                                                 - characteristic fluid dynamic time
                                                                                      	au_{\mathrm{fluid}}
                                                                                                     (=V_C^{1/3}u^{-1}),
           - temperature, [K]
                                                                                                  - maximum recycle factor due
\hat{T}_1
           - preheating oven temperature, [K]
                                                                                                     to aerodynamic configuration, [-]
           - fluid velocity, [ms<sup>-1</sup>]
                                                                                                     chemical destruction rate of
           - computational cell volume, [m<sup>3</sup>]
                                                                                                     H_2O_2, [mol·m<sup>-3</sup>s<sup>-1</sup>]

H<sub>2</sub>O<sub>2</sub> mass fraction, [-]
hydrogen mass fraction in mixture i, [-]
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