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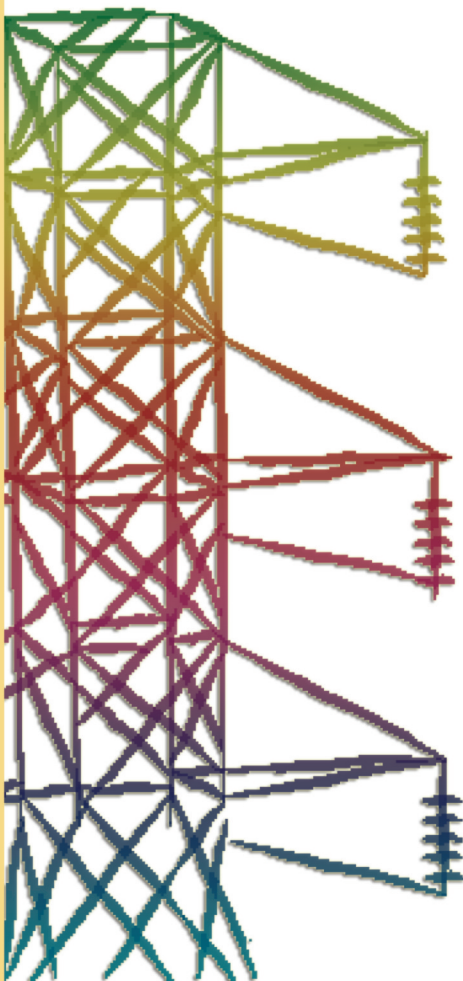
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Effects of Hot Diluted Oxidant Flow on Reactive Structures in HDDI MILD Combustion

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1. Introduction

The typical structure of not premixed diffusion flames in standard conditions can be significantly modified whether injected flows are diluted and/or pre-heated.

The increase of the fuel and/or oxidant flow dilution up to extreme conditions could lead to the formation of mixtures not ignitable so that the oxidation processes could be sustained just in case the pre-heating temperature of one of the two flows is high enough to promote the auto-ignition of the system. The flows high initial enthalpy and the low fuel and/or oxygen concentration can drastically modify the structure of the oxidative region as well as the physical and chemical kinetics respect to conventional diffusion flame. Such operative conditions are typical of Mild combustion processes [1]. More specifically a combination of both heating and dilution of oxidant and/or fuel yields a not-premixed combustion process which is named Hot Diluted Diffusion Ignition (HDDI) [2, 4] when

•   heating contributes significantly to the creation of an oxidative structure in the sense that no combustion process occurs without it;

•   dilution is so intense that the maximum temperature attainable inside this structure is so low that, in turn, it affects significantly its placement in the mixture fraction domain, the structure itself and the physical and chemical kinetics when compared to a diffusion flame process.

Four combinations of opposed jets configurations main parameters are possible:
1.   Heating of oxidant   and   dilution of fuel
2.   Heating of oxidant   and   dilution of oxidant
3.   Heating of fuel   and   dilution of fuel
4.   Heating of fuel   and   dilution of oxidant.

In particular in this work the sub-categories HODO (Hot Oxidant Diluted Oxidant), corresponding to condition 2, is considered.

2. Numerical tools

The two interacting hot and/or diluted flows have been schematized by means of opposed jets. In particular, among the possible systems that can be considered on the basis of the flows characteristics the attention has been focused on the configuration schematized in Figure 1.

Oxidant flow is composed by oxygen and nitrogen with an inlet temperature T_{in}. The dilution level is expressed by the O_2 molar concentration. The fuel flow is composed by methane with an inlet temperature T_O and an inlet velocity V_o. The oxidant flow velocity comes from the assumption that the kinetic energies of the two flows are set equal so that the stagnation point is located, for any feed conditions, in the middle of the two injection planes, 2 cm apart. The asymptotic strain rate (K_O) is defined with respect to the fuel flow velocity as $K_O = V_0 / D$.

The numerical analysis was carried out by means of Oppdif [5] application of ChemKin [6] package by using the GRI 3.0 [7] oxidation mechanism. Oppdif computes the diffusion flame
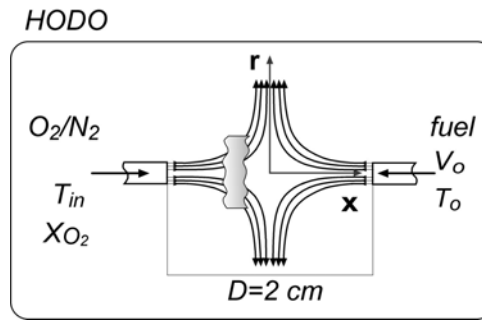
between two opposed nozzles.

Fig. 1   HODO configuration.

The structure of reactive zone was analyzed by a simple evaluation of temperature T and heat release rate *H* profiles as a function of the mixture fraction Z along the axial coordinate of the system.

In the case of standard inlet conditions, corresponding to undiluted jets, fed at $T_{in} = T_o = 300$ K, the profiles of these quantities are reported in Figure 2 as a function of mixture fraction with solid and dashed lines, respectively. The fuel flow, composed by un-diluted methane, is fed with an inlet velocity equal to 25 cm/s. The system pressure is set equal to 10 bar.
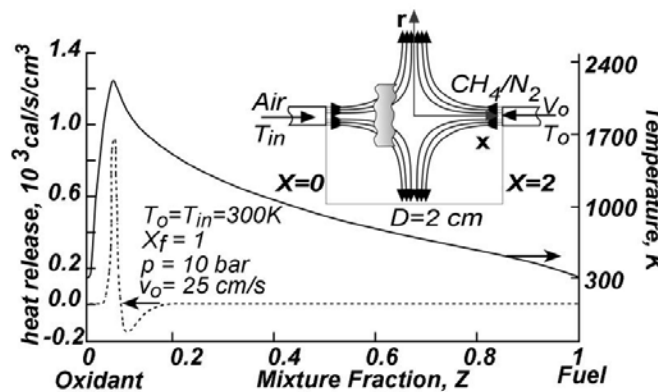

Fig. 2   Temperature (solid line) and heat release rate (dashed line) versus mixture fraction, at $T_o=T_{in} = 300K$, $X_f=1$ and p = 10bar.

Starting from $T_{in} = 300$ K at Z = 0 the temperature rapidly increases up to the maximum value of about 2220 K, occurring at the stoichiometric value of the mixture fraction (Z_{st}). Hereafter, T smoothly decreases along Z to reach T_o at Z = 1 (i.e. in correspondence with fuel inlet section at x = 2 cm). The related *H* profile, reported in the same figure with a dashed line, better highlights the range of Z where reactions take place. Starting from Z = 0, *H* sharply increases up to a maximum that occurs in correspondence with the maximum temperature, at $Z_{st} = 0.055$. This trend testifies a prevalent reaction activity toward the formation of species with heat of formation lower than that of reactants, i.e. intermediate or product of oxidation reactions. Then *H* drops toward a negative minimum, which occurs at about Z = 0.08. Later on it becomes zero and keeps this value up to Z = 1. In contrast with what occurs where *H* is positive, the range of Z corresponding to the negative *H* values identifies the region where pyrolytic reactions overcome oxidation ones.

Several simulations were performed over a wide range of T_{in} at different dilution level of fuel jet and strain rate K_o in adiabatic conditions at low and high pressure. In particular, for lack of space, the results reported in this chapter are relative to systems at 10 bar, but they are in

agreement with the ones obtained at atmospheric pressure.

3. Numerical Results

In order to understand the modifications of the oxidative structure induced by the high dilution degrees and high pre-heating temperatures the system schematically reported in Figure 1 has been considered. The numerical simulations have been carried on varying X_{O2}, from air composition up to very diluted conditions as well as the pre-heating temperature of the oxidant flow.
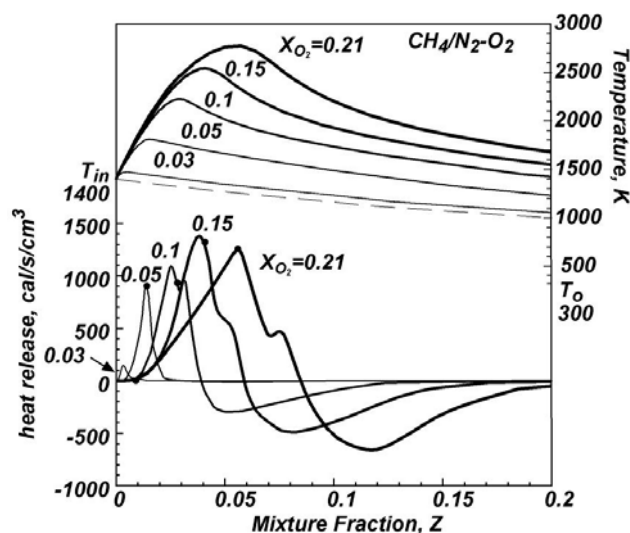

Fig. 3   Temperature (upper part) and heat release rate (lower part) versus mixture fraction, parametric on X_{O2} at T_o= 300K, T_{in} = 1400K and p = 10bar.

Figure 3 shows the temperature and the heat release profiles along the coordinate Z on curves parametric in the dilution level of the N_2/O_2 pre-heated flow. T_{in} is equal to 1400K, while the pressure is 10 bar. Methane is fed at environmental temperature, whilst its inlet velocity V_o is 100 m/s. It corresponds to an asymptotic strain rate K_o equal to 50 s^{-1}. Thickness of solid lines decreases with X_{O2}. They represent T and H, whilst dots indicate, the stoichiometric values of the parameter Z as X_{O2} changes.

Since the reactive region in the Z coordinate develops in the neighborhood of the oxidant injection plane the analysis of the results has been restricted in the range 0 - 0.2 of the mixture fraction.

The profile at X_{O2}= 0.21 corresponds to the case where the oxidant flow is air. In such system the temperature slightly increases up to reach a maximum value equal to 2750 K, then it slowly decreases down to 300 K towards the methane injection plane.

At the same time, the heat release shows two relative maxima. The first, in correspondence of the maximum value of temperature, located at $Z = Z_{st} = 0.055$, the latter, with a lower value, at $Z = 0.06$. The heat release shows a minimum negative value at $Z = 0.1$. Afterwards it slowly increases up to zero towards the fuel injection plane.

As soon as the nitrogen content in the oxidant flow is enhanced, the maxima of the temperature and the heat release profiles occur for lower values of Z and the region, in the Z range, where reactions occur is narrower with respect to the case with no oxidant dilution.

For $X_{O2} = 0.15$ the maximum temperature (T_{max}= 2675 K) occurs for at $Z = Z_{st} = 0.04$. The absolute maximum of H is localized at the same position, while the second one at $Z = 0.50$. Both the two maxima heat release values are higher than the ones showed in the previous case, whereas the negative minimum is lower in absolute value.

When the oxidant stream dilution degree is 90% in nitrogen (X_{O2}=0.1), the enhancement of the heat capacity of the system leads to a lower system temperature. The maximum allowed temperature is positioned at $Z = 0.0275$ which again corresponds to Z_{st}.

The heat release profile is similar to the previous cases, although the absolute maximum is not located in correspondence of Z_{st}. As matter of a fact the first maximum is sited at $Z = 0.025$, whereas the second one at $Z = 0.031$. In such a case the two maxima values are of comparable values, furthermore the negative heat release region is reduced and the minimum absolute value is diminished in comparison with the previous condition.

A further depletion of oxygen causes a merging of the heat release maxima values and almost a disappearance of the pyrolitic region. As matter of fact in case of $X_{O2} = 0.05$ the H profile shows a single positive maximum in correspondence of the maximum system temperature for $Z = Z_{st}$, while the negative region is negligible.

The last case reported on the diagram of Figure 3 refers to the molar fraction of oxygen equal to 0.03. For such a dilution the temperature increase is very limited and the maximum temperature is 1475 K, while the heat release presents one single maximum very low in comparison with other cases. Neither the maximum temperature, nor the maximum heat release are correlated with Z_{st}.

The heat release profile shapes have been considered as indicative of the structure of the oxidation region. In particular profiles that show two positive peaks and a negative minimum value are indicative of high temperature combustion, where the composition of the mixture ensures high heat release and high system temperatures so that the oxidation process is self-sustaining. On the other hand, heat release profiles that present a single positive peak and no negative values are symptomatic of Mild Combustion. In such case oxidation reactions occur just by means of the high enthalpy content provided by the hot oxidant flow.

On the basis of the numerical heat release combustion, the numerical results have been reassumed in a map of behavior T_{in}-ΔT reported in Figure 4 for a pressure equal to 10 bar and a strain rate equal to 50 s^{-1}. Four different regions, respectively named "no combustion", "feedback combustion", high temperature combustion" and "Mild HDDI" are identifiable on the plane on the basis of heat release profiles.

The region "no combustion" establishes in correspondence of inlet temperatures lower than the auto-ignition temperature of a stoichiometric mixture and for oxidant flow compositions very diluted so that the heat release associated to combustion is not sufficient to sustain the oxidation process.

Where the composition of the mixture can satisfy the last requirement, the oxidation process can evolve through traditional feed-back structures.

An inlet temperature increase causes the shifts towards the "high temperature combustion" region where both the pre-heating temperature and the mixture compositions also contribute to the oxidation reactions, but they do not change the main features of the diffusive structure.

The last region is related to Mild-HDDI, where the high pre-heating oxidant flow temperature promotes and sustains the combustion process. It is identifiable for T_{in} comprised between 1200K and 1500K and ΔT from 0 up to 800 K.

It is worth noting that the crossing from the "feedback combustion" to the "high temperature" regimes does happen through a transitional region where two different system steady states are allowed. This indicates a hysteresis behavior from low to high methane conversion solutions.

At the same time a transitional region is identifiable also for the passage from "no combustion" to "mild combustion" but numerical results show just a single solution indicating the disappearance of the hysteresis with a continuous transition from low to high fuel conversions.

The analysis relative to species heat release has been performed also for counter-flows flames

where a pre-heated and diluted nitrogen-oxidant flow is fed towards methane at environmental temperature. In particular Figure 5a shows the heat release distribution in the coordinate Z for a system in which the oxidant flow is not diluted and is pre-heated up to 1400K. The pressure is 10 bar and the strain rate is 50 s^{-1}.
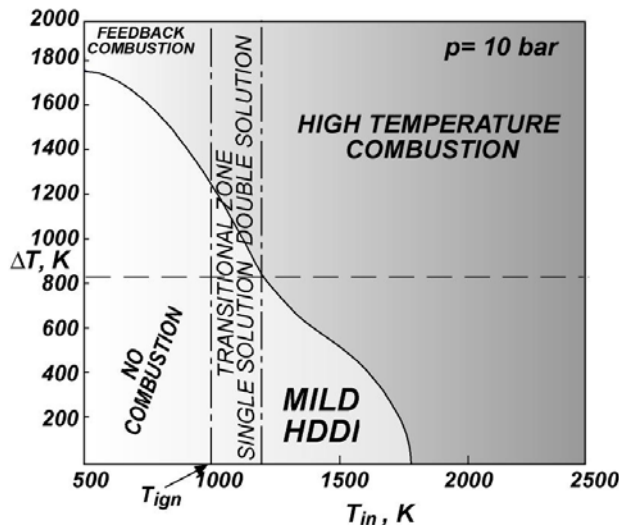

Fig. 4   Combustion regimes at p = 10bar, T_o = 300K, T_{in} = 1400K and K_o = 50 s^{-1}.

In the map of behavior such conditions fall in the High Temperature Combustion region, since no dilution and high inlet temperature are considered. The global heat release, reported in gray, has been widely described in the same section. The global *H* maximum value occurs in correspondence of Z_{st}, where OH, CO and CO_2 exhibit their maxima. Reactions relative to production and consumption of OH give the highest contribution to the *H* magnitude. The second global *H* peak is mainly correlated to CH_3, CO_2 and C_2H_2 species, while CO and OH give a negative contribution. C_2H_2 mainly contributes to the negative region for higher values of Z. The structure of the oxidative region appears to be very complex and several pathways are identifiable during the oxidation of CH_4.
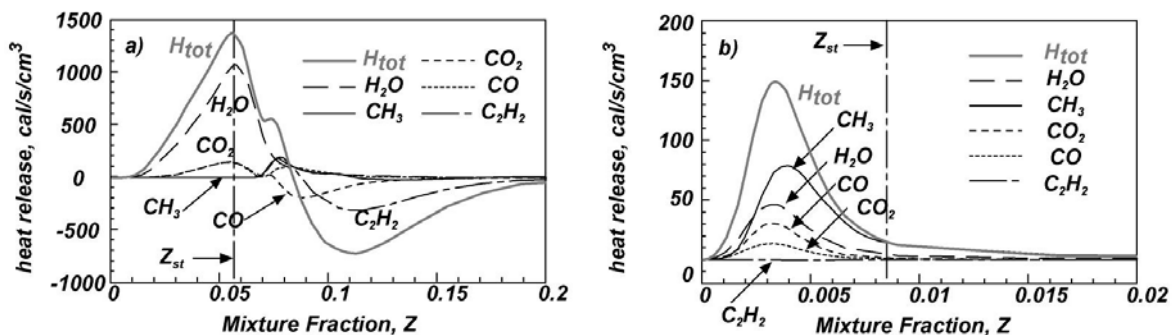

Fig. 5   Global heat release relative and heat release relative to selected species at T_{in}=1400K and (a) X_{O2}=0.21 and (b) X_{O2}=0.03.

The heat release shown in Figure 5b is indicative of Mild Combustion regime. The global heat release shows a single peak that does not take place at the stoichiometric mixture fraction and no pyrolytic reaction occurs. The global heat release as well as the heat release relative to OH, CO and CO_2 present their maximum at Z = 0.03, while the methyl radical at Z slightly higher than the other curves (Z = 0.034).

The trend of curves suggest that a simpler and more uniform structure of the oxidative region in Mild operative conditions is obtained.

4. Conclusions

The numerical results highlght a drastic change of the structure of the reactive region in HODO systems with respect to traditional diffusion flame configuration. In particular the main effects of dilution and pre-heating of oxidant flow on the kinetics of methane oxidation can be reassumed:

1.   The location of maximum heat release does not take place at the stoichiometric condition, but it occurs in a lean condition where frozen mixture temperature is higher than the self-ignition of the stoichiometric mixture and it contributes significantly to the reactant temperature. This is counter-intuitive for combustion engineers because their understanding is always related to diffusion flames without heating and dilution so that they think it is a basic rule of a diffusion controlled process that maximum heat release takes place at the stoichiometric conditions.

2.   The heat release is distributed in a wide range of the mixture fraction domain. This is counter-intuitive for combustion engineers because their understanding is related to diffusion flames without heating and dilution so that the flame structure is basically thought to be "infinitely thin" or "thin".

3.   The maximum local temperature of the systems is lower than that of undiluted diffusion flames even though one of the reactant is pre-heated. This lowers all the temperatures and in particular those related to rich conditions, which are also diluted, so that pyrolytic kinetics are much slower than undiluted not-heated conditions. This yields, in turn, the disappearance of a distinct pyrolytic region where heavy molecular weight carbonaceous and nitrogen substituted products are generated. The lower temperature affects also the oxidative routes by favoring oxygen addition on partially reformed fuel in respect to oxidation of thermally decomposed species.

4.   The temperature is more uniformly distributed along the mixture fraction space with respect to undiluted not-heated conditions. This is due to the shift of the stoichiometric condition toward higher values of the mixture fraction and the simultaneous decrease of the product temperature at stoichiometric condition. In other words the reactant (in this case the oxidant) temperature and the flame adiabatic temperature are more similar when both preheating and dilution increase. The maximum allowable temperature between these two states is linearly interpolated so that all the temperatures are more uniform.

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