Thermal Flame Structure of Laminar Methane/H₂/Air Mixtures. Influence of Reactants Preheating

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1. Introduction

The combustion of fossil energy leads to significant pollution and is probably one of the major contributing factor in global warming. Lean premixed combustion is one of the most promising concepts for reduction of pollutant emissions. But this solution leads to flame instabilities. One way of avoiding the flames instabilities is to avoid extinction phenomena (Bradley et al., 1998). Hydrogen addition seems to be a promising solution. Understanding the fuel composition impact on combustor performance or emissions requires the understanding of the mixture fundamental combustion properties. Data and studies concerning the laminar flames description can be found in numerous papers and experimental data concerning diluted or enriched methane/air laminar flame speed can be found in the literature (Ilbas et al., 2006; Law et al., 2004; Schefer, 2003). But hardly any experimental data concerning thermal structure of such flames can be found. Knowing the thermal laminar H₂-enriched methane-air flame structure should help to understand the mechanism leading to flame stability increase or pollutant emission reduction. This paper is mainly dedicated to an experimental and numerical investigation of the effect of reactants preheating on H₂-enriched methane-air inner flame structure.

2. Experimental setup and data processing

2.1. Experimental set-up

The experimental setup consists of a vertical wind tunnel (Renou et al., 2004) with a mean flow velocity of 4m/s. The fuel-air mixture is directed to a divergent-convergent wind tunnel and is laminarized with a honeycomb structure. At the exit, a V-shaped flame is stabilized on a heated rod. Since the flame is laminar, the investigated flame conditions are only defined by H₂ percentage (in volume), reactants temperatures and equivalence ratios. The 2D Rayleigh imaging system is mainly composed of a tripled 10Hz frequency Nd-YAG laser (355nm, 220mJ/pulse). An optical arrangement enables a laser sheet to be obtained in the study zone, chosen over 30mm from the burner exit to avoid stretch effects. The Rayleigh signal images are collected with an ICCD camera (284*386 pixels² Lavision Flamestar), gated at 200ns. The spatial resolution of the whole optical collection system has been measured and is approximately 260µm (Lafay et al., 2008).

2.2. Data reduction

The computation method of $T(x,y)$ applied to our results is based on a computed flame library. The originality of the method is the use of a Rayleigh scattering signal-based progress variable, which allow us to deduce the temperature from the only information available, i.e. the Rayleigh Scattering Signal (RSS) (Lafay et al., 2008). The flame thickness definition used in this study is:

$$\delta_j^o = \frac{T_{\text{max}} - T_{\text{min}}}{(\nabla T)_{\text{max}}}$$

(1)
The validity and the accuracy of both the experimental data and the data post-processing have been checked. It has been performed with methane-air flames at ambient temperature and with reactants preheated at 480K. First, we present images and RSS profiles of two methane-air flames (Figure 1) with the reactants preheated at 480K. Since RSS is a function of $T^{-1}$, the highly preheated flames images are expected to be the most shot-noise affected.

![Figure 1](image1)

**Fig. 1** Raw normalized images of RSS (left) and corresponding raw profiles across the flame front (right). Reactants temperature is 480K. Profiles are plotted in flame coordinates.

The RMS of the Rayleigh signal (and thus of all the derived quantities) is the highest beyond the Maximum Temperature Gradient (MTG), in burnt gases. On this figure, images are corrected following the procedure described in (Lafay et al., 2008). To avoid shot to shot variations effects, the profiles are normalized by their fresh gases mean values. It must be noted that neither image nor profile have been filtered. Secondly, in order to validate the measurement of temperature and of temperature gradient, the flame thicknesses of numerous ambient temperature and preheated methane-air flames are compared to the numerical computed flame thicknesses. These latest have been computed by using the Cantera software package to solve the classical set of steady ODEs (Kee et al., 2003), leading to the computation of the adiabatic laminar flame speed. The GRImech3.0 has been used to close this set of equation. Finally, the complete assessment of the measurements is performed by comparing the temperature gradient across the flame front to the numerical one.

### 3. Results and discussion

#### 3.1. Influence of preheating on methane air flames

The influence of reactants preheating on laminar flame thickness is presented Figure 2. Both experimental and numerical results are discussed. First, the numerical predictions agree well with the measurements: the difference between them is less than the error bars, which represents the flame thickness RMS (10%). The disagreement at high $\phi$ and high preheating temperature is due to the limited spatial resolution of the optical system. This section is also devoted to the assessment of our temperature measurement, both for the MTG and for the temperature gradient. The progress variable $c$ is defined as follow:

$$ c = \frac{T - T_u}{T_b - T_u} $$$$ c \text{ unburnt gases temperature} $$$$ c \text{ burnt gases temperature} \tag{2}$$

The plot of $\nabla c$, $\langle \nabla c \rangle$ and $\nabla \langle c \rangle$ versus $c$ for 480K methane-air flames are presented Figure 2. To minimize noise in these data, 10 profiles of the progress variable are averaged (Knaus et al., 2005) to give what we consider to be an instantaneous profile.
A summary of preheating temperature influence is also presented Figure 2. It is shown that the temperature gradients in the preheat zone are strongly affected when the lean flame fresh gases are preheated. First, the observable inflection point at $c = 0.05$ for the 300K flame is progressively attenuated when the preheating temperature increases. In this zone, the gradient is small and evolves with a quasi-linear shape. None chemical reactions are observable and the heat transfer by conduction induced by the heat release occurring at higher $c$ is predominant. Secondly, the location of $\nabla <c >_{\text{max}}$ is progressively and slightly shifted toward lower $c$ (from 0.65 to 0.57). Its value is obviously increased. The post flame region ($c > c_{\nabla <c >_{\text{max}}}$) is not affected by preheating. The reactant temperature seems to strongly influences transport and chemical (mainly) phenomenon from the preheat zone to $c = c_{\nabla <c >_{\text{max}}}$.

All these observations demonstrate the ability of both the experimental setup and the data reduction procedure to measure the flame thickness, but also to measure the temperature gradient in the flame front.

### 3.2. Results of H$_2$ enriched and preheated methane-air flames

Figure 3 shows the experimental flame thicknesses of the %40 H$_2$ enriched methane-air flames (reactants temperature from 300K to 480K). For the 300K methane-air flames, the discrepancies between numerical and experimental flame thicknesses (Figure 3) appear with $\phi$ lower than 0.55 (Lafay et al., 2008). For these conditions, the experimental results give lower values than the numerical simulations. It shows that the chemical mechanism (GRI Mech 3.0) fails to predict the MTG for ultra lean high H$_2$-enriched methane-air flames. Concerning the flame thickness with reactants preheated from 350 to 480K, the numerical and experimental results still disagree. The experimental results show a moderate increase of flame thickness when $\phi$ decreases, whereas numerical values increase drastically. The Figure 3 shows the numerical/experimental results ratio. This ratio is close to unity for the richest flames, and rapidly increases for $\phi <0.5$. Increasing the preheating temperature at a constant $\phi$ leads to a decrease of the numerical-experimental disagreement. Thus, the chemical mechanism is not able to predict the experimental MTG, but its sensibility to temperature reduces the discrepancy.
Fig. 3 Influence of preheat temperature on %40 H₂ enriched methane-air flame thickness (left). Numerical-experimental flame thickness ratio (left).

Fig. 4 Plot of $\nabla c$, $\langle \nabla c \rangle$ and $\nabla \langle c \rangle$ versus $c$ for a %40 H₂ enriched methane air flames and for different reactants preheating temperature.

Concerning the preheated flames structure, Figure 4 shows the plot of $\nabla c$ and profile of $\langle \nabla c \rangle$ and $\nabla \langle c \rangle$ versus $c$ for a 300K, 350K, 420K and 480K %40H₂ enriched methane air flames ($\phi=0.45$). At highest temperature, numerical and experimental $\langle \nabla c \rangle_{\text{max}}$ agree, and the predictions are accurate in the preheat zone. As the reactants temperature decreases, the numerical results show a overestimated decrease of the $\nabla c$. As previously described, the lowest temperature (300K) at $\phi=0.45$ shows a great disagreement between numerical and experimental data both in the preheat zone and around $\langle \nabla c \rangle_{\text{max}}$. 
Fig. 5: Evolution of $\nabla <c>$ versus $c$ with H$_2$ enrichment and preheat temperature

The evolution of $\nabla <c>$ versus $c$ when the reactants temperature of the $40\%$H$_2$ enriched methane-air flame is increased is plotted Figure 5.

First, we can observe that H$_2$ enrichment strongly (at T=300K) shifts the value of $\nabla <c>_{\text{max}}$ toward lower $c$ (from 0.75 to 0.5), but doesn’t markedly change the “shape” of the global curve: an inflection point still remains at $c \approx 0.05$ and the slope of the curve is almost constant in the preheat zone (only a slight shift toward higher values can be observed). With the definition of $c$ given in the previous section, we have:

$$c|_{\nabla <c>_{\text{max}}} = \frac{T_{b} - T_{u}}{T_{b} - T_{u}}$$

At constant $\phi$, the investigated change in H$_2$ concentration doesn’t strongly modify the temperature jump (i.e. $T_{b} - T_{u}$) across the flame front: only a maximum difference of 30K can be observed both on the measurements and on the computation. Thus, a decrease of $c|_{\nabla <c>_{\text{max}}}$ implies a decrease of $T_{b} - T_{u}$, due to both a higher heat diffusion rate (hydrogen diffusion) and a higher chemical mixture reactivity at low temperature (Dagaut et al., 2005; Lafay et al., 2008).

Second, following the arguments given previously for hydrogen enrichment, and since the temperature jump across the preheated flame front is not markedly changed, we can say that the reactants preheating, surprisingly, only shifts the temperature at the MTG toward higher value: $T_{b} - T_{u}$ is almost a constant. This unexpected increase of $T_{b} - T_{u}$ shows that the mixture reactivity at low temperature is not enhanced with increasing reactants temperature. The temperature at the MTG seems not to be driven by reactants temperature, but mainly by radicals that are produced in the reaction zone and back diffuse upstream.

4. Conclusion

The effects of temperature on H$_2$ enriched laminar methane-air flame inner thermal structure have been studied experimentally using the Rayleigh scattering technique applied on a V-shaped flame. First, the effect on temperature on pure methane-air flame thermal structure has been carried out both to assess the measurement quality and to set a reference for further comparisons. The experimental pure methane-air flame results are compared to numerical ones and are found to be in a good agreement for all the operating conditions. The first effects of the reactant preheating are the reduction of the flame thickness and a extent of the flammability domain. Secondly, for both the methane-air and the H$_2$ enriched methane-air...
flames, the reactant preheating is found to strongly increase the temperature gradients in the preheat zone. Moreover, the slope of the curve $\nabla c$ versus $c$ in the preheat zone becomes steeper with reactants preheating, whereas it is not markedly changed in the post flame region $c > c_{\text{post}}$. Finally, for the $40\%$ H$_2$ enriched methane-air flame, the reactants preheating doesn’t strongly shift the value of $c_{\text{preheat}}$ toward lower $c$. It means that, at the opposite of hydrogen enrichment which shows lower $c_{\text{preheat}}$ with increasing hydrogen proportions, the preheating of hydrogen-enriched methane-air flames doesn’t change the relative position of the maximum temperature gradient in the flame front.

5. References