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Laminar flame speed evaluation for CH_4/O_2 mixtures at high pressure and temperature for rocket engine applications

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Abstract

Pure methane-oxygen mixtures in liquid rocket engines lead to extreme pressure and temperature conditions that are prohibitive for most of the experimental setups. Hence, there is very little data on such flames in the literature, especially concerning the laminar flame speed S_u, often limited at atmospheric pressure. The recent development of methalox rocket engines, which design process often requires CFD calculations, brings this lack of data to the forefront. Indeed, the CFD simulations require valid chemical schemes in the real operating conditions. To address this problem, flame measurements have been performed in a special isochoric combustion chamber with full optical access (OPTIPRIME) developed at ICARE. An extensive database in conditions never tested before is generated for several equivalence ratios, temperature and pressure ranges. Multiple chemical mechanisms were then compared to those results, showing various levels of agreement. Hence, the best mechanism from the literature on OPTIPRIME results and other literature experimental data was selected. A sensitivity analysis was performed to identify key chemical reactions controlling the flame speed. These key reactions could later be tuned by an optimization process to perfectly match the experimental results. Finally, additional measurements were performed in order to develop a S_u=f(P,T) correlation to build a future flame speed database under rocket engines relevant conditions.

Keywords: Rocket engines; Methane-oxygen; High-Pressure; Laminar flame speed; Flame kinetics

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1. Introduction

With the space market being more and more competitive, reusable launchers are envisioned as a key technology to reduce the costs of access to orbit. Encompassed in this logic, methane (CH_4) has been identified as the best candidate fuel for the development of low-cost reusable space launch systems (high energy density, low operating cost, and good overall performances in terms of specific impulse [1]). As a consequence, several methane-based projects are currently being studied and developed around the world [2][3], making the understanding of pure methane/oxygen combustion (flame dynamics, kinetics, etc.) at high pressure and temperature a key problematic. Hence, available kinetic models need to be confronted to reliable experimental data.

However, the high reactivity of methane with pure oxygen (O_2) generally prevents its experimental study whether it is ignition delay, species profiles, or flame speed measurements. Indeed, such mixtures release a significant amount of energy compared to classically studied CH_4/air mixtures where nitrogen (N_2) acts as a thermal ballast. Therefore, the resulting temperature of burnt gases is very high (> 3000 K against 2100 K in air at equivalence ratio φ = 1 and 1 bar) leading to high laminar flame speeds S_u (around 3 m.s^{-1} against 0.3 m.s^{-1} in air under the same T, P conditions). Therefore, bibliographical information on this topic is very scarce.

As diffusion flames in rocket engine relevant conditions require very complex and expensive dedicated setups, premixed laminar flame speeds are more straightforward to measure. They are a fundamental parameter to study and can be used to assess the behavior of kinetic mechanisms [4]. Furthermore, it has been proven that a chemical mechanism valid for premixed flame configurations is generally also valid for stretched laminar diffusion flames. Indeed, simplified kinetic models validated at equilibrium and in premixed laminar flames show a similar behavior to diffusion-flames-derived chemistry in LES calculations of rocket engine configurations [3]**.** Nevertheless, as mentioned above, the extreme conditions of rocket engine combustion chambers are generally incompatible with most of the experimental devices used to measure flame speeds. Therefore, diluted mixtures using an inert gas as a thermal ballast (such as N_2, Ar or He) have been much more investigated. In order to study CH_4/O_2 flames, another option is to focus on very lean or very rich conditions where S_u values are lower.

To the best of our knowledge, very few studies were performed in CH_4/O_2 mixtures. The first are related by Lewis and Von Elbe [5]. More recently, measures were performed by Mazas et al.[6][7]. Both studies used a Bunsen-type burner as experimental setup. In Mazas case, a low diameter nozzle (3 mm) was used to reduce the turbulence effect on the flame. Flame speeds were measured thanks to a Schlieren technique for different equivalence ratios φ varying from 0.5 to 1.6 at atmospheric pressure and 298 K. Additional measurements were carried out at atmospheric pressure for lean, stoichiometric and rich mixtures, varying the initial temperature of the reactive mixture from 298 K to 500 K. However, no pressure variation was considered in this study, nor in the work of Lewis and Von Elbe. Therefore, extending the database to a wider range of temperatures and pressures is greatly valuable in order to develop a combustion model for CH_4/O_2 mixtures under rocket engine conditions.

The current study provides data acquired with the perfectly spherical and isochoric combustion chamber (OPTIPRIME) with full optical access developed at ICARE [8]. φ was varied from 1 to 2.5, with a pressure range depending on the tested conditions going from 0.3 to 18 bar while the fresh gases temperature vary between 298 and 603 K (and the burnt gases temperature from 2260 K to 3115 K). The obtained database, complemented with other measurements from the literature, is then used to build a valid kinetic model for methane oxycombustion at high pressure.

2. Experimental data acquisition

To be as close as possible to the conditions of interest, high pressures and temperatures are targeted. This is not straightforward as can be seen in the literature, where methane flame speeds are mostly studied around atmospheric pressure and low temperatures for technical reasons [9]. In order to go to higher pressures, the isochoric combustion method is used (also referred as spherically expanding flame at constant volume). The principle is to record the evolutions of pressure and flame radius over time and compute the fresh gases temperature assuming an isentropic compression, allowing to evaluate the flame speeds under these conditions. In OPTIPRIME, these simultaneous measurements are made possible thanks to an optical access with a 360° fused silica ring. The flame speed determination process has been detailed in previous publications [8][4]. Hence, the chamber allows to measure S_u values from ambient (or even sub-atmospheric) conditions up to 20 bar at high temperatures (up to 603 K in the fresh gases, 3115 K in the burnt gases). It is important to note that CH_4/O_2 mixtures lead in the studied conditions to very high flame speeds near stoichiometry (up to

almost 6 m.s^{-1}), which is 10 times higher than the values encountered for diluted mixtures previously studied with OPTIPRIME. Such strong combustion induces an important mechanical and thermal stresses on the setup as well as challenging conditions for the data acquisition and post-treatment. These challenges require to perform several checks and adaptations which are detailed below.

2.1 *Pressure effects*

The pressure time-variation can be very steep for CH_4/O_2 mixtures. At φ = 1.5 for example, there is a factor of 30 between the dP/dt encountered with CH_4/Air (400 bar.s^{-1}) and CH_4/O_2 (12000 bar.s^{-1}). In order to withstand these brutal variations at each firing, the 360° silica ring used for optical access has a thickness of 11 mm. However, in order to avoid destructive levels at the end of the flame propagation where the pressure is at its maximum, the initial pressure P_0 was sub-atmospheric at 0.5 bar for most of the cases. This also allows to delay the occurrence of hydrodynamic instabilities (visually detectable) providing longer S_u recorded signals. On the other hand, lower initial pressure leads to slightly higher flame speed than at atmospheric pressure. Hence, the acquisition frequencies of the camera and pressure transducers need to be increased (up to respectively 110000 fps and 60 kHz). This requires an adaptation of the data filtering parameters for the post-treatment compared to what was previously used for CH_4/air mixtures.

2.2 *Stretch effects*

Another monitored parameter is the flame stretch. Numerical computations of flame speeds were performed for non-stretched flames. In order to compare the calculated values to the experimental ones, stretch dependence needs to be assessed. In spherical flames, the stretch is limited to curvature and writes as $\kappa = 2/R_f \, . dR_f/dt$. The high values of flame speed encountered for CH_4/O_2 mixtures then lead to high stretch values. Typically, in diluted cases, the initial κ is far lower than in non-diluted mixtures by a factor 10. In the present case, κ_{ini} is found around 400 s^{-1} for CH_4/air where it is 4000 s^{-1} for CH_4/O_2 at stoichiometry. These stretch levels were evaluated when R_f is large enough to start computing S_u. In practice in CH_4/air mixtures, the instant when the flame is considered not to be affected by stretch corresponds to the time when the pressure reaches 2 times its initial value P_0 [8]. This relation is based on the Markstein length in the burnt gases L_b, computed from experimental data, assuming the linear relation $S_b = S_b^0 - L_b\kappa$ (S_b^0 being the unstretched flame speed in the burnt gases). The criterion marks the time when $L_b\kappa \ll S_b^0$ (or very low values of the $L_b\kappa/S_b^0$ ratio), i-e the stretch effects are negligible. In view of the observed high κ values, this evaluation needs to be conducted for CH_4/O_2 mixtures. In order to assess the evolution of the different parameters over the whole flame propagation process inside the chamber, the $L_b\kappa/S_b$ ratio is preferably used, while L_b is supposed constant over pressure and temperature [10]. In order to cover the whole range of studied equivalence ratios, $L_b\kappa/S_b$ is studied for φ=0.5, $\varphi = 1$ and φ=2.5. As illustrated in Table 1, κ_{ini} values for the first recorded R_f vary with the equivalence ratio φ, κ strongly decreasing as φ tends toward richer mixtures since the flame speed diminishes. Concerning lean mixtures, the stretch progressively increases toward stoichiometry, following the flame speed evolution.

Table 1
Computed Markstein length and critical stretch values from experimental data at different φ for CH_4/O_2 mixtures

φ (−)	κ_{ini} (s^{-1})	L_b (m)
0.5	2000	2.72E-04
1	4000	1.60E-04
2.5	400	8.09E-04

Furthermore, it can be noticed that the Markstein length L_b is greater for rich and lean mixtures than for stoichiometric ones. Since this parameter roughly reflects the flame front response to an exterior perturbation, which is here the stretch κ, it means that rich and lean CH_4/O_2 flames tend to have a higher sensitivity to stretch than stoichiometric ones. This is indeed observed on Fig 1, illustrating the $L_b\kappa/S_b$ evolution over R_f for the studied cases.

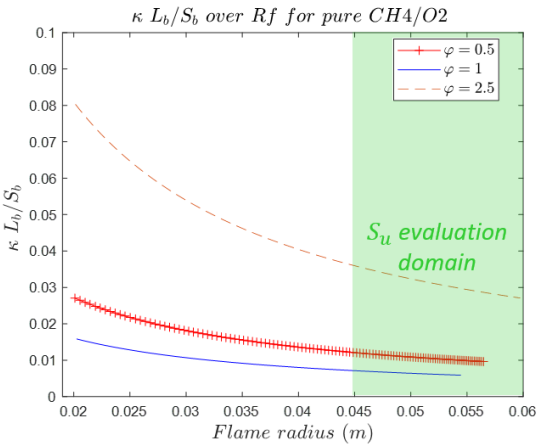


Fig. 1: $\kappa L_b/S_b$ ratio over flame radius R_f for 2 different equivalence ratios (CH_4/O_2 mixture)

The figure confirms that stretch effects are stronger for rich and lean mixtures, despite the lower κ values. However, as stated earlier, the evaluation of S_u starts when the pressure has reached $2P_0$ (region labelled as "evaluation domain" in Fig. 1). This threshold is reached for substantially similar flame radii for the studied conditions. It occurs at the end of flame

propagation [8], i-e at low stretch levels. Indeed, $L_b\kappa/S_b$ is lower than 5% for both studied mixtures in the measurement zone. Thus, the pressure criterion is still valid to determine the first R_f to be considered for flame speed calculation in CH_4/O_2 mixtures.

3. Comparison with numerical kinetic mechanisms

The chemical mechanisms investigated in this paper (cf. Table 2) are from various sources. Some were designed on purpose for high-pressure methane/oxygen applications such as RAMEC [11], Slavinskaya [12] and Zhukov [13]. RAMEC and Zhukov are mainly validated on the basis of ignition delay times, the first one using low dilution ratios conditions at high pressure. Slavinskaya mechanism is validated on both high-pressure diluted ignition delay times and laminar flame speeds in the air at atmospheric pressure. The others selected models are multi-purpose mechanisms like GRI-Mech3.0 [14], FFCM1 [15] and POLIMI C1-C3 [16]. They were validated on various targets, including flame speeds, but at pressure and temperature far from rocket engine relevant conditions. FFCM1 and GRI-Mech 3.0 are compact mechanisms, allowing fast calculations. FFCM1 previously shown satisfactory results on OPTIPRIME CH_4/air experiments [17]. POLIMI C1-C3 requires more computation time but was validated on numerous experimental targets.

The mechanisms were used in 1D laminar flame computations performed with the PREMIX code from CHEMKIN Pro [18] at various P, T and φ conditions identical to the experiments.

Table 2
Tested chemical kinetic mechanisms

Mechanism	#Species	#Reactions
Slavinskaya[11]	22	97
Zhukov [12]	23	51
RAMEC [10]	38	190
FFCM1 [14]	38	291
GRI-Mech 3.0 [13]	53	325
POLIMI C1-C3 [15]	114	1999

First, an evaluation of these mechanisms against the experimental dataset of Mazas was performed at constant pressure (1 bar) and temperature (298 K). The overall agreement is good while some mechanisms (RAMEC in particular) underestimate S_u as φ increases. Others, like FFCM1, constantly underestimate the experimental values. When compared to Mazas parametrical study on initial temperature T_0 at 1 bar, all mechanisms except POLIMI C1-C3 show some discrepancies with the experiment as T_0 increases, for all tested φ (the figures can be found in the supplementary materials – Figs S2 to S4). Hence, it is now interesting to compare the mechanisms with the experiment at varying P and T.

Experimental results from φ = 0.5 to 2.5 are displayed in Fig 2. All tests started at P_0 = 0.4 bar except the case φ = 2.5 at P_0=2 bar for which S_u is lower. S_u is represented as a function of P (right side of each graph) and T (left side) with the associated experimental uncertainty of $\pm$5%. The flame speed increases with simultaneous augmentation of P and T as the fresh gases are compressed by the flame front. Maximum usable pressure goes from 1.5 to 18 bar depending on the considered equivalence ratio. All the experimental data can be found in the supplementary materials. Fresh gas temperature ranges between 300 and 603 K while the burnt gases temperature varies from 2260 to 3115 K. Unstretched laminar flame speeds S_u^0 (i-e isobaric S_u) determined by using the zero-stretch extrapolation method described in section 2 are also indicated on Fig 2. The measured flame speed is maximum at stoichiometry between 450 and 550 cm.s^{-1} and then decreases with increasing φ.

Numerical results also reported in Fig 2 show that all tested mechanisms dramatically underestimate the experimental flame speed values for lean, stoichiometric and near-stoichiometric conditions. At φ = 2, most of the mechanisms are close to the measured values while they all over-estimate them at φ = 2.5. It is important to note that the last two equivalence ratios are beyond the range over which most of the mechanisms have been validated. However, all mechanisms well reproduce the evolution shape of S_u, i-e its bending when P and T increase. This bending effect seems to be more important at low pressure (as seen at φ=2), confirming the trends suggested by the isobaric S_u^0 and the flame speed traces for most of the tested conditions. On average over all φ conditions, POLIMI C1-C3 is found to best predict experimental results, with a relative error varying between 26 and -10% depending on the tested conditions. Interestingly, FFCM1 behaves as an offset of POLIMI C1-C3, always underestimating S_u but mimicking well its evolution over P and T. All other mechanisms, apart from specific cases, remain in between POLIMI C1-

C3 and FFCM1. Hence, it seems all the mechanisms initially tailored for methane oxycombustion fail to capture the right S_u levels. For RAMEC and Zhukov, this may be due to the fact that their experimental validation is only based on ignition delay times. For Slavinskaya, the S_u targets in the air are too far from oxycombustion conditions. As for FFCM1 and GRI-Mech 3.0, although some of the numerous S_u validation targets are at high pressure, the important dilution rate leads to lower temperatures than CH_4/O_2 mixtures. Hence the observed discrepancies. Moreover, many of the sensitive (i-e important) reactions depicted in the next section are defined differently from one mechanism to another.

Therefore, POLIMI C1-C3 seems to be the best starting point for a yet-to-be-optimized mechanism versatile for both CH_4/air and CH_4/O_2, as well as highly diluted mixtures. Indeed, other data acquired at high pressure (up to 20 bar) for stoichiometric highly diluted mixtures (up to 80% diluent in molar mass in the oxidizing mixture, diluent is composed of 70%He and 30%Ar, presented in the supplementary materials) confirm the rather good agreement obtained with POLIMI C1-C3 for these conditions. Overall a major outcome of this study is the lack of validity of existing mechanisms for CH_4/O_2 at high P due to the lack of data, which is partly overcome by the present OPTIPRIME measurements.

4. Sensitivity analysis of the POLIMI C1-C3 mechanism

As seen in the previous section, the calculations performed with POLIMI are in closest agreement with our experimental data. However, this mechanism is not able to fully reproduce the S_u behavior for CH_4/O_2 mixtures in the targeted (P,T,φ) conditions. In order to better understand the kinetics under these conditions and identify the rate constants, k, on which it would be possible to act to obtain a better agreement, a sensitivity analysis of S_u were performed for different φ (from 0.5 to 2.5) for the P and T range covered during the experiment. The

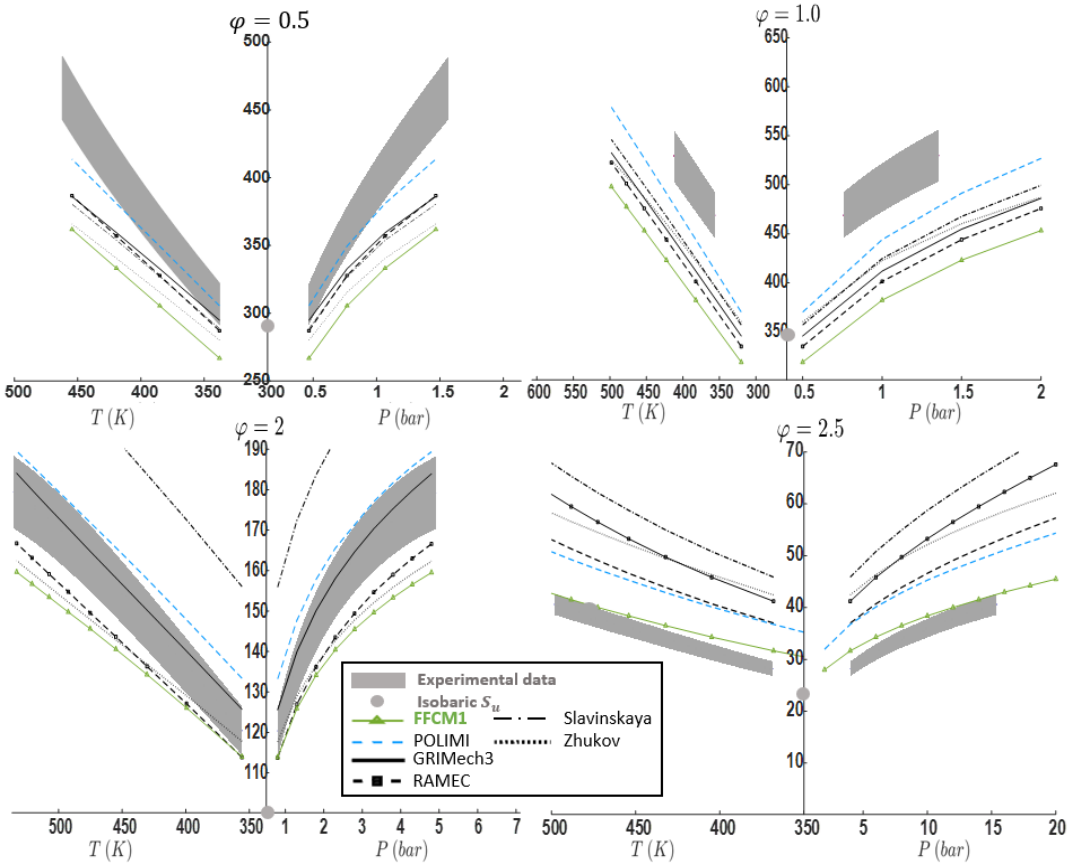


Fig. 2: Flame speed S_u [cm.s^{-1}] for CH_4/O_2 mixtures at various equivalence ratios φ (thickened traces) as a function of T and P compared to several kinetic mechanisms (lines). Single symbols are for experimental isobaric flame speeds.

sensitivity is defined as $S = \partial S_u/\partial k_j$ where k_j is the rate constant of reaction j. A panel of 13 most sensitive reactions was identified and displayed on Fig 3.

These driving reactions for the flame speed decompose in 2 families: those which have a promoting effect on S_u ($S > 0$) and those which have an inhibiting effect ($S < 0$). For all conditions, $H + O_2 = O + OH$ is by far the most sensitive reaction, its S increasing with φ. For other $S > 0$ reactions, $HCO + M = H + CO + M$ and $OH + CH_3 = H + CH_2OH$ seem non-negligible whatever the equivalence ratio. Interestingly $OH + CO = H + CO_2$ is quite sensitive at lean conditions while quickly becoming almost insensitive when φ increases. In addition, $H + CH_4 = H_2 + CH_3$ is the most important $S < 0$ reaction, with growing sensitivity as the equivalence ratio increases. Its value is only exceeded by $O_2 + HCO = HO_2 + CO$ for lean conditions only. $H + HCO = H_2 + CO$ also plays a significant role with a sensitivity not varying much which φ. The reaction $O + CH_3 = H + CH_2O$ however, has almost no impact at lean and stoichiometric conditions but becomes the second largest negative-sensitive reaction at φ = 2. Interestingly, the sensitivity sign of $OH + CH_4 = H_2O + CH_3$ changes with φ. Being positive for lean conditions (since it adds CH_3 in an environment where there is only a few), it gradually transits toward negative values for reach conditions

All these reactions also play a role for the tested diluted mixtures mentioned before. On the other hand, $CH_3 + H(+M) = CH_4(+M)$, identified as a key reaction for CH_4/air flame speeds [4] is far less sensitive for oxy-combustion. Thus, it does not play an important (though not negligible) role here. However, complementary sensitivity analyses conducted at higher pressures for CH_4/O_2 tend to show that its S increases with P, which makes the additional role of high pressure diluted experimental measurements where this reaction is also sensitive more important for a future optimization process. This work is currently in progress, using the OptiSMOKE++ [19] code to tune the Arrhenius parameters of several sensitive reactions identified here.

5. Flame speed correlation

As seen in the previous sections, a single firing with the OPTIPRIME setup allows to access a full trace of flame speeds S_u over a variety of (P, T) conditions. Therefore, it is interesting to perform several firings, varying the initial pressure in order to have different traces allowing to explore other domains. It is then possible to fit a S_u= f(P, T) correlation on the obtained data, allowing to interpolate in between the traces and even extrapolate outside the measurement range, creating a full map limiting the number of experiments. This method would allow to progressively build a flame speed database for several mixtures, temperature and pressure conditions that could be used in a CFD combustion model. Such a work is currently underway at ICARE for CH_4/air mixtures.

In order to cover a broader range of conditions, additional firings are performed for the same set of equivalence ratios as previously tested at an initial pressure of 0.5 bar (P_0 = 3 bar for φ = 2.5). It is important to mention that the margin to choose the conditions to interpolate and extrapolate from is very thin for these extreme CH_4/O_2 mixtures compared to

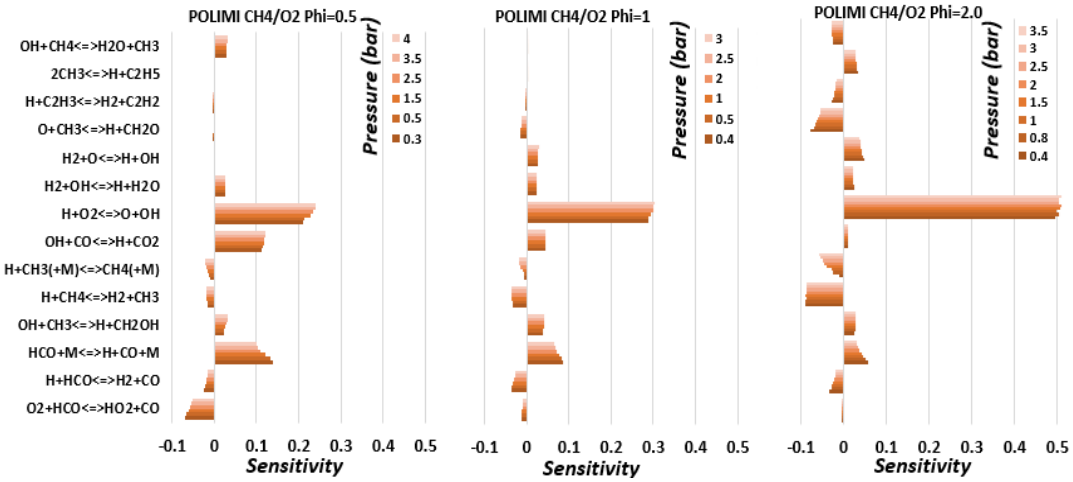


Fig. 3: Sensitivity analysis on S_u of POLIMI C1-C3 for a CH_4/O_2 mixture at different equivalence ratios φ

the diluted mixtures (for example with air), because of the limiting pressure derivatives values reached during the flame propagation in OPTIPRIME. Obtained experimental S_u traces are displayed in the supplementary materials in Fig S5. In order to enhance the model precision, data from Mazas at atmospheric pressure presented in the first section of the manuscript are added as target values for the correlation.

The first S_u= f(P, T) correlation used in this study to fit each equivalence ratio condition is described by Hu et al. [20][21]. It is based on the initial flame speed S_{u0} at reference conditions P_0, T_0. Two exponents, α_T and β_P, respectively functions of the temperature and the pressure contain five different coefficients that can be tuned to fit the experimental traces.

$$S_u = S_{u0}(T/T_0)^{\alpha_T}(P/P_0)^{\beta_P} \quad (1)$$

Obtained numerical values of the function parameters are given in the supplementary materials. A different fit is generated for each φ conditions. Hence there is as many maps as tested equivalence ratios. Fig 4 illustrates an example of a correlation map for a CH_4/O_2 mixture at φ = 2.5. The experimental traces are represented in black, while the rest of the map is either interpolated or extrapolated within the range of the covered pressure and temperatures. This kind of map allows to predict the evolution of S_u over pressure and temperature, the map being more precise when new traces are added.

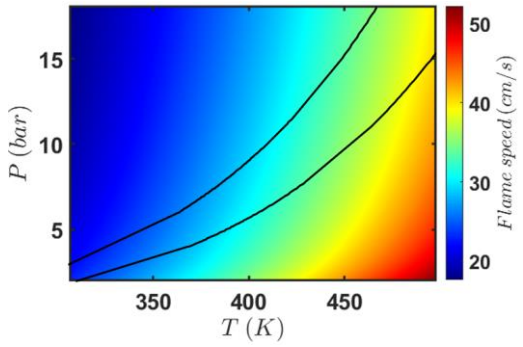


Fig. 4: S_u=f(P,T) correlation map at φ =2.5

Finally, the obtained correlation was compared to Mazas T_0 parametric database (graphs are available in the supplementary materials), showing satisfactory results. The models stay inside the range of experimental uncertainty for each φ condition tested by Mazas.

In order to simplify the modelling, another approach considering the equivalence ratio dependency was used. It consists of an expression similar to Equation 1, but with S_{u0}, α and β function of equivalence ratio φ. Such expression is described in the paper of Wu et al. [22]. This approach allows the use of a single correlation for all the range of applicable (P, T, φ). The results, also shown in the supplementary materials, were compared to Mazas $S_u = f(\varphi)$ study at 1 bar. Fitting is quite good for lean mixture and stoichiometry while it slightly overestimates the experience for rich conditions. The observation is identical on Mazas T_0 parametric database.

6. Conclusion

The development of methane-oxygen rocket engines for reusable launch vehicles requires an extensive knowledge of the chemical kinetics in extreme pressure and temperature conditions. However, it is precisely those hard-to-reach conditions that are prohibitive for experimental test benches, leading to very limited experimental flame speed datasets in the literature, often restricted to atmospheric pressure. In order to bring additional knowledge on the flame speed behavior at high P and T for those mixtures, the OPTIPRIME isochoric combustion chamber with full optical access was used. Measurements were performed for a variety of equivalence ratios (from 0.5 to 2.5) and a large range of pressures, depending on the tested cases (up to 1.5 to 18 bar). Kinetic mechanisms, for most of them traditionally used to describe CH_4/O_2 combustion are compared to OPTIPRIME and literature datasets. It allows to point out major discrepancies between the numerical and experimental results when it comes to extreme conditions. Finally, the POLIMI C1-C3 model seems to be the best candidate for a yet-to-be optimized mechanism to describe CH_4/O_2 combustion. Several reactions were identified as key ones that need to be better evaluated. In addition, S_u=f(P, T) correlations were built from OPTIPRIME results in order to create a full map of flame speed for diverse conditions. Additional experiments are still required to increase accuracy and extend the range of validity.

Declaration of competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary materials are available for this article.

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