

# Characterization of combustion in small volumes

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## Abstract

A preliminary estimation of combustion characteristics in micro combustors is performed with the Perfectly Stirred Reactor model. This model allows us to qualitatively assess the range of stable combustion, as well as the extinction limits for different fuels, inlet temperatures, equivalence ratios and heat loss rates. Linked to low turbulence rates, the weak mixing effects on chemistry are particularly studied with the Partially Stirred Reactor model. Three micromixing models (Vulis, Interaction by Exchange with the Mean and  $\beta$ -PDF) are compared and the crucial influence of mixing at molecular scale on combustion extinction limits is pointed out. Then, practical cases with estimated operating conditions are tested and results are discussed.

## 1 Introduction

Micro scale engines for power generation and air vehicle propulsion applications have received considerable attention during the last decade. We can note, for example, several new concepts of micro-machines as micro gas turbine [1], toroidal “Swiss Roll” counterflow heat exchanger [2] or micro thermoelectric converter [3]. However, scale-down of macro-scale combustion devices implies new problems concerning flame stabilization and combustion efficiency. Various technical difficulties, which may not be significant or well known in macro scale combustors, become crucial for combustion in small volumes ( $< 1\text{cm}^3$ ). The most specific difficulties are : small mean residence time ( $\sim 1\text{ms}$ ) which is comparable with chemical times (Damköhler number  $\sim 1$ ); weak turbulent mixing linked to modest Reynolds number ( $\text{Re} \sim 500$ ); and significant heat losses on walls due to large ratio between combustor surface and volume [4]. A theoretical analysis of combustion in small volumes has to be led to design an adapted geometry of micro combustor and prepare future experiments.

## 2 Specific Objectives

In order to get a preliminary estimation of the low Damköhler number influence on the overall characteristics (extinction limits, combustion efficiency, mean temperature), a simplified approach is used to model the combustion chamber. At first, we suppose that the mixing inside the combustor is instantaneous. Then, the Perfectly Stirred Reactor (PSR) model, widespread in the combustion community [5], is considered to perform a parametric study on relevant parameters (temperature of inlet gases, type of fuel, equivalence ratio, heat losses rate). Practical cases are tested for relevant operating conditions. Those conditions are close to the MIT micro turbine configuration [1].

Due to the very small residence time in combustor, variations linked to non-perfect turbulent mixing become considerable and play a prominent role on combustion. To study these turbulence-chemistry interactions, the Partially Stirred Reactor (PaSR) model [6] is used : inside the combustion chambers, the fluid is broken up into discrete fragments or “pockets” with a very small size and these fragments are uniformly dispersed throughout the reactor. The molecular exchange between pockets is controlled by micromixing. In most models of turbulent reactive flows currently used, the chemical production is treated exactly while micromixing influence has to be modeled. In this study, three different approaches of micromixing modelisation are studied and compared : Interaction by Exchange with the Mean (IEM) model [8]; Vulis model [7]; and assumed  $\beta$ -PDF model [9, 10]. These models of molecular mixing modify the finite rate chemistry used in our calculations. In a next step, the

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most appropriate micromixing model will be included in a CFD code for quantitative calculations of combustion in small volumes.

### 3 Modelisation

#### 3.1 Perfectly Stirred Reactor model

First of all, to understand the effect of small Damköhler number on combustion characteristics, the Perfectly Stirred Reactor model is used. The combustor is modeled by an open reactor in which inlet gases are instantaneously well mixed after injection. Mixing between fresh and burnt gases is perfect for all turbulence scales. That is, composition, temperature and density are considered uniform throughout the reactor. In this model, mean values of temperature and concentrations coincide with instantaneous ones. The inlet gas is a premixed air-fuel reactive mixture and different ratios between hydrogen and methane are considered in this study. A detailed chemical kinetic mechanism is used for the air-H<sub>2</sub>-CH<sub>4</sub> combustion with 79 reactions and 21 species [11]. The instantaneous production rate  $W$  of species  $n$  is given by the law of mass action

$$W_n = M_n \sum_{r=1}^{N_r} \left[ (\nu''_{nr} - \nu'_{nr}) \left( k_{fr} \prod_{i=1}^{N_s+1} C_i^{\nu'_{ir}} - k_{br} \prod_{i=1}^{N_s+1} C_i^{\nu''_{ir}} \right) \right] ,$$

where  $M_n$  is the molecular mass of species  $n$ ,  $N_r$  the number of reactions involved,  $k_{fr}$  and  $k_{br}$  are the reaction rates of reaction  $r$ ,  $\nu'_{ir}$  and  $\nu''_{ir}$  are stoichiometric coefficients, and  $C_i$  ( $i = 1, \dots, N_s$ ) is the concentration of species  $i$ . The last concentration  $C_{N_s+1}$  represents third-body species and  $t_{i,r}$  the third-body efficiency for reaction  $r$  (these last coefficients are fixed to unity in our calculations)

$$C_{N_s+1,r} = \sum_{i=1}^{N_s} t_{ir} \frac{\rho Y_i}{M_i} . \quad (1)$$

For stationary state, the classical balance equations for species, enthalpy and mass conservation are

$$\frac{Y_{0,i} - Y_i}{\tau_r} = - \frac{W_i(Y_i, T)}{\rho} , \quad (2)$$

$$\frac{h_0 - h}{\tau_r} = \frac{Q}{\rho} , \quad (3)$$

$$\tau_r = \frac{\rho V}{\dot{m}} , \quad (4)$$

where  $Y_i$  and  $Y_{0,i}$  are the reactor and the inflow mass fraction of species  $i$ ,  $\tau_r$  is the mean residence time,  $\rho$  the density,  $h$  and  $h_0$  are the reactor and the inflow mass enthalpy,  $Q$  is the heat loss rate,  $V$  the volume of combustor and  $\dot{m}$  the mass flow rate. The mean residence time inside the combustor depends on the mass flow rate, the pressure of gases and the volume of reactor. Eqn. 3 respects the enthalpy conservation between the system inlet and outlet for an adiabatic case ( $Q = 0$ ). In the non-adiabatic case, heat losses are modeled by a linear function of the sensible enthalpy creation during combustion

$$Q = \frac{A}{\tau_r} \int_{T_0}^T Cp(T) dT , \quad 0 \leq A < 1 , \quad (5)$$

$$Cp(T) = \sum_{i=1}^n Y_i Cp_i(T) , \quad (6)$$

where  $C_p$  is the specific heat of the mixture at constant pressure and  $A$  is the heat loss coefficient. Thermodynamic properties and mass fractions of the reactive mixture are determined for a stationary state and a fixed residence time. For extinction limit study, calculations are started with an ignited combustion and a high residence time in comparison with the chemical time. Then, the balance equations are solved for a stationary state with smaller and smaller residence times until a combustion extinction is observed. This critical residence time is compared for different conditions of inlet temperature, fuel composition, equivalence ratio, and heat loss rates previously presented. All results of this parametric study are presented and discussed in section 4.

### 3.2 Partially Stirred Reactor models

Partially Stirred Reactor (PaSR) models have been developed to take into account finite micromixing times  $\tau_m$ , that is, the presence of scalar fluctuations at the molecular scale inside the combustor. First developed by Correa [6], this model have been used previously by Sabel'nikov and Figueira da Silva to show the role played by the ratios between the residence time and the micromixing time and chemical times, respectively on combustion [12]. Fluctuations of temperature and species concentrations have a great influence on the highly non-linear term  $W_i$  presented in Eqn. 1. The three models presented in present paragraph take into account micromixing influence on chemical productions. For simplicity, heat losses are not treated for the PaSR case and total enthalpy is kept constant for stationary conditions.

#### 3.2.1 Interaction by Exchange with the Mean model

The IEM model introduced by [8] is based on a lagrangian point of view and takes into account an age repartition of fluid particles inside the combustor for the micromixing modeling. Injected fluid particles are mixed with older ones in burnt pockets of gases. Furthermore, mixing between all particles depends on the micromixing time scale  $\tau_m$ . The instantaneous mass fraction values in the reactor  $Y_i(t)$  for each given residence time  $\tau$  follow the equation

$$\frac{dY_i}{dt} = \frac{\bar{Y}_i - Y_i}{\tau_m} + \frac{W_i(Y_i, T)}{\bar{\rho}} \quad , \quad 0 < t < \tau \quad , \quad Y_i(t=0) = Y_{i,0} \quad . \quad (7)$$

The mean concentration  $\bar{Y}_i$  is the integral of  $Y_i$  weighted by the PDF  $p(Y_i)$  which is representative of the species concentrations repartition in the reactor

$$\bar{Y}_i = \int_0^1 Y_i p(Y_i) dY_i \quad . \quad (8)$$

When the combustion is stationary, an ergodicity assumption is made between the concentration repartition for a given time and the particle age repartition inside combustor

$$p(Y_i) dY_i = f(t) dt \quad , \quad (9)$$

and thus Eqn. 8 yields

$$\bar{Y}_i = \int_0^\infty Y_i(t) f(t) dt \quad . \quad (10)$$

The particle age PDF  $f(t)$  corresponds to a Poisson distribution : particles are randomly added to the reactor while other ones are randomly removed. This repartition has a characteristic form described by  $\frac{e^{-t/\tau_r}}{\tau_r}$ , where  $\tau_r$  is the mean residence time.

For each instantaneous composition, the corresponding instantaneous temperature is determined from the total enthalpy balance equation. The mean temperature inside the reactor is calculated by integration of all instantaneous temperatures with the ergodicity assumption and a particle repartition following a Poisson distribution.

### 3.2.2 Vulis model

The Vulis model [7] is a global and much simpler approach than the IEM model presented in the previous paragraph. In this model, the impact of micro-mixing is taken into account by introducing in the chemical production rates calculation some “virtual” concentration  $Y_{v,i}$  different from the mean concentration  $\bar{Y}_i$ . The species balance equations are determined for a stationary state with a modified production term from the PSR case.

$$\frac{\bar{Y}_i - Y_{i,0}}{\tau_r} = \frac{W_i(Y_{v,i}, T)}{\bar{\rho}} \quad . \quad (11)$$

Furthermore, Vulis has proposed to close the previous equation by defining the virtual mass fractions  $Y_{v,i}$  from an additional balance equation based on micromixing time and production rates,

$$\frac{Y_{v,i} - \bar{Y}_i}{\tau_m} = \frac{W_i(Y_{v,i}, T)}{\bar{\rho}} \quad . \quad (12)$$

If we define  $x$  as the dimensionless ratio between  $\tau_r$  and  $\tau_m$ , the combination of Eqn. 11 and 12 leads to the following relation between “virtual” and mean mass fractions inside the reactor

$$Y_{v,i} = \bar{Y}_i \left( 1 + \frac{1}{x} \right) - \frac{1}{x} Y_{i,0} \quad , \quad x = \frac{\tau_r}{\tau_m} \quad . \quad (13)$$

Consequently, when the micromixing time is negligible compared with the residence time, the virtual fractions tend to the value of mean mass fractions and results for stationary state are the same as in the PSR case. Conversely, when  $x$  is small, the virtual fractions,  $Y_{v,i}$ , tend iteratively to the inflow values,  $Y_{i,0}$ , and production terms are calculated for a composition fixed to  $Y_{i,0}$ . Consequently, if the inlet gas temperature is not sufficient to ignite combustion, for a high micromixing time, the stationary solution is a slow combustion regime.

### 3.2.3 Assumed PDF approach

In order to model the influence of weak mixing on combustion, fluctuations of temperature and species concentrations are evaluated with a statistical approach. The concentration balance equation is solved with an average chemical production and for a stationary state. The mean production rate of species  $n$  can be determined from :

$$\bar{W}_n = \int_0^{+\infty} \int_0^1 W_n P(T, C_1, \dots, C_{N_s}) dT dC_1 \dots dC_{N_s} \quad (14)$$

In order to simplify the integration of the above equation, in most studies, the density is kept constant at its averaged value and a statistical independence of temperature and gas composition is assumed. Thus, the PDF  $P$  can be written as :

$$P(T, C_n) = P_T(T) P_Y(Y_1, \dots, Y_{N_s}) \delta(\bar{\rho} - \rho) \quad (15)$$

These assumptions permit to decompose the integration of Eq. 1 in two statistically independent parts. On one side, the reaction rate depends of the temperature and, on the other side, the concentration product depends of the species mass fractions. For those two terms, PDF shape are assumed :  $\beta$ -PDFs for concentrations distributions and a clipped gaussian PDF shape is used for the temperature repartition.

### Multivariate $\beta$ -PDF of composition

First introduced by Richardson et al. [10], the  $\beta$ -PDF is commonly used to model the concentration fluctuations in turbulent flows. In fact, this type of assumed PDF is able to handle fluctuations which are not symmetric about the concentration mean value, as well as allow bi-modal behaviour for the early stage of mixing. Furthermore, we will see in this paragraph that  $\beta$ -PDF offers convenient mathematical aspects and can be easily integrated. This  $\beta$ -PDF approach has been extended to multiple scalar mixing by Girimaji [9] in order to apply this model to full kinetic schemes. Thus, for  $N_s$  species, the joint PDF is

$$P_Y(Y_1, \dots, Y_{N_s}) = \frac{\Gamma\left(\sum_{i=1}^{N_s} \beta_i\right)}{\prod_{i=1}^{N_s} \Gamma(\beta_i)} \left[ \delta\left(1 - \sum_{i=1}^{N_s} Y_i\right) \prod_{i=1}^{N_s} Y_i^{\beta_i-1} \right] ,$$

where

$$\beta_n = \overline{Y}_n \left[ \frac{\sum_{i=1}^{N_s} \overline{Y}_i (1 - \overline{Y}_i)}{\sigma_Y^2} - 1 \right] , \quad \beta_n \geq 0 \quad \text{and} \quad \sigma_Y^2 = \sum_{i=1}^{N_s} \overline{Y}_i'^2 .$$

The multi-variate  $\beta$ -PDF is completely defined given the mean mass fractions and the sum of mass fraction variances ( $\sigma_Y^2$ ). An attractive feature of this type of assumed PDF is that only one transport equation for  $\sigma_Y^2$  is required regardless of the number of composition scalars being considered. Furthermore, thanks to convenient mathematical aspects, the integration of the production term described by Eq. 1 can be done analytically. For the reaction  $r$ ,

$$\overline{\prod_{i=1}^{N_s+1} C_i^{\nu_{ir}}} = \prod_{i=1}^{N_s} \left( \frac{\overline{p}}{M_i} \right)^{\nu_{ir}} \frac{\prod_{i=1}^{N_s} \prod_{k=1}^{\nu_{kr}} (\beta_i + k - 1)}{\prod_{k=1}^{\sum_{m=1}^{N_s} \nu_{mr}} \left[ \left( \sum_{i=1}^{N_s} \beta_i \right) + k - 1 \right]} \left[ \sum_{j=1}^{N_s} t_{jr} \frac{\overline{p}}{M_j} \frac{(\beta_j + \nu_{jr})}{\sum_{k=1}^{N_s} (\beta_k + \nu_{kr})} \right]^{\nu_{N_s+1r}} .$$

If the third-body plays a role in the reaction considered, the  $\nu_{N_s+1r}$  coefficient is equal to one. If not, this coefficient stays to zero. Finally, an exact solution of the chemical production rate is available for a low CPU cost which increases linearly whatever the number of reactive species considered in the chemical scheme.

### Gaussian PDF of temperature

To model the effects of temperature fluctuations on the chemical production rate defined in Eq. 1, an assumed Gaussian PDF  $P_T(T)$  is used [15]. Based on physical considerations, an assumed PDF of temperature is defined. The temperature fluctuations must stay between two physical limits,  $T_0$  the inlet temperature and  $T_{ad}$ , the adiabatic flame temperature based on local conditions. The unwanted “tails” of the distribution in chosen as the ranges  $T < T_0$  and  $T > T_{ad}$  are replaced by two Dirac delta functions at  $T_0$  and  $T_{ad}$ , respectively [17]. The PDF may be expressed as

$$P_T(T) = \frac{1}{\sqrt{2\pi\sigma_c^2}} \exp \left[ -\frac{(T - T_c)^2}{2\sigma_c^2} \right] [H(T - T_0) - H(T - T_{ad})] + A_0 \delta(T - T_0) + A_{ad} \delta(T - T_{ad}) ,$$

where  $H$  is the Heaviside function and  $A_0$  and  $A_{ad}$  are the areas of clipped parts.  $T_c$  and  $\sigma_c^2$  correspond to corrected values of temperature and variance in agreement with PDF normalisation properties for first and second moment. As previously, for the mass fractions variance, a transport equation with micro mixing model for the enthalpy variance is solved.

## 4 Results and Discussion

### 4.1 Parametric study with the PSR model

Several parameters having influence on combustion in small volumes are studied in ranges corresponding to possible operating conditions for a micro power generator. This parametric study represents a data base obtained with a full kinetic chemistry mechanism and shows the respective importance of each parameter on combustion extinction limits. Those data should be compared with more accurate calculations results of the complete combustor geometry performed with a CFD code.

Figure 1 a. shows the temperature evolution inside the reactor according to different residence

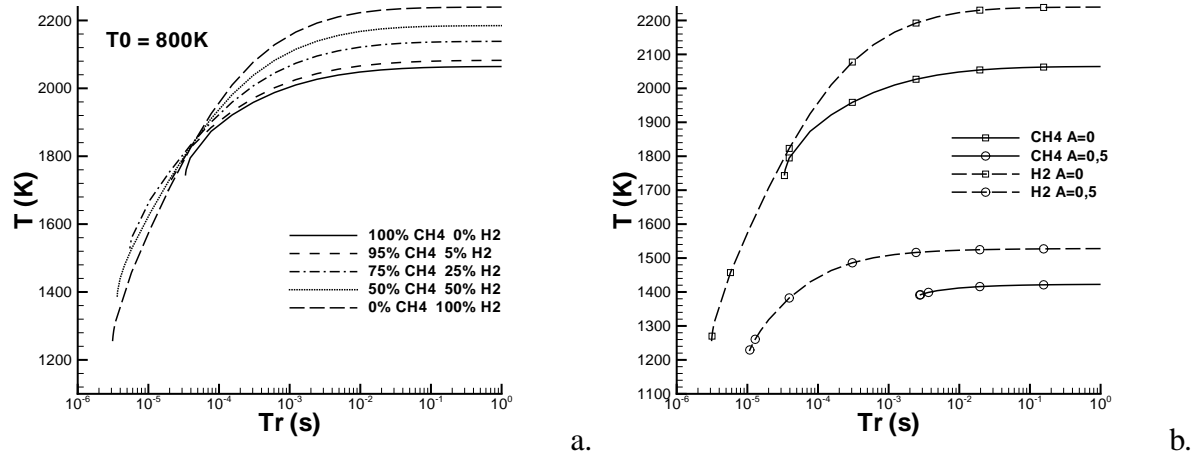


Figure 1: Evolution of temperature for different fuels (a.) and for different heat loss coefficients (b.)

times, from one second to extinction limit, for several fuel compositions. Those results are obtained for an inlet gases temperature of 800K and with adiabatic conditions. We can notice that the critical residence time of extinction strongly depends on the fuel properties. Those extinction limits are determined for several temperatures from 700K to 1000K of injected gases and it appears that inlet temperature plays a great role on combustion extinction limits. Thus, the critical time can be divided by two (from  $5 \times 10^{-5}$ s to  $2.5 \times 10^{-5}$ s for methane) when the inlet temperature increases by 300K. As expected, a hydrogen-air mixture has a smaller critical residence time than a methane-air mixture. We can notice that only 5% of hydrogen added to methane allows to divide by two the critical residence time of the reactive mixture. Moreover, a Norton and Vlachos study [16] shows that the propane-air combustion is more robust than the methane-air one. Propane could be a good compromise between combustion stability, observed with hydrogen, and storage facilities offered by hydrocarbon fuels. Another important parameter is the equivalence ratio of the inlet reactive mixture. For our operating conditions, the change of the equivalence ratio from 0.5 to 0.8 can lead to multiply the critical residence time by three. The temperature level of combustion decreases strongly with the equivalence ratio for both fuels studied. Furthermore, a difference of 400K on stationary state temperature is observed in the range considered.

Figures 1 b. show the crucial influence of wall heat transfers on combustion temperature and extinction critical time. A high heat loss rate leads to a strong decrease of the temperature level, the combustion efficiency and then the global efficiency of the micro system. In order to limit those transfers, a thermal insulation of combustor walls can be imagined. In the case of the MIT micro turbine, heat losses have been estimated in a range of 5% to 10% with a maximum value of 30% in more unfavourable cases [4]. The critical residence time increases dangerously for a methane-air

combustion and reaches the same order of magnitude as the characteristic residence time of micro systems ( $\tau_r = 5 \times 10^{-4} s$ , [1]), and thus, the flame stability is threatened. Finally, we can notice that the hydrogen combustion is less affected by heat losses than the methane one.

## 4.2 Micromixing models comparison and PaSR model

The three micromixing models are tested with realistic reference conditions close to real case. Results are exposed on figure 2 a., for the methane-air combustion, and figure 2 b., for the hydrogen-air combustion. First of all, we can notice that the assumed PDF model gives very different results than

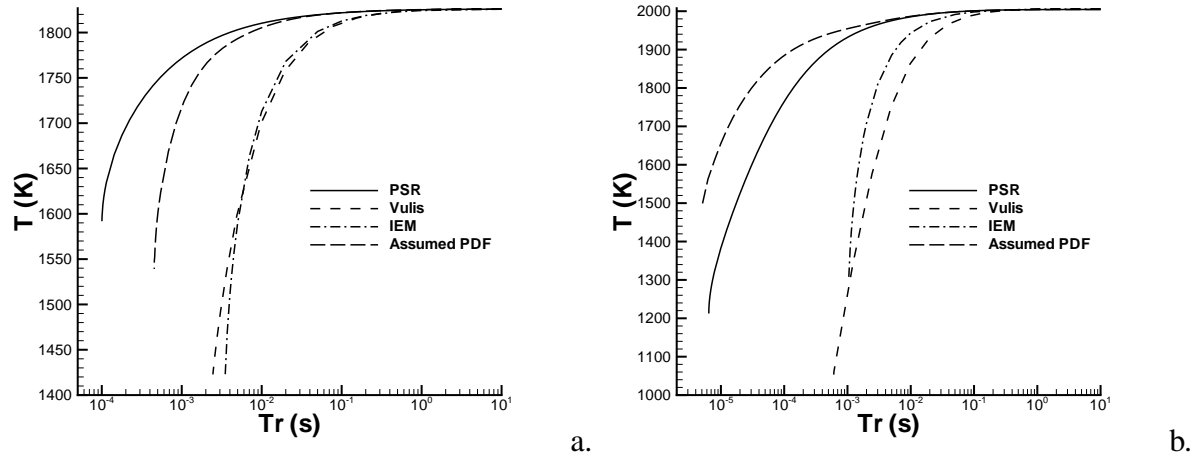


Figure 2: Extinction limits for a methane-air (a.) and for a hydrogen-air mixture (b.),  $\tau_m = 10^{-3} s$

the others micromixing models. Then, several physical inconsistencies can be noted for this approach. In fact, multi-variate  $\beta$ -PDF shape is determined for each species by a “global variance”  $\sigma_Y^2$ . This quantity connects fluctuation of each species together. The physical analysis of the evolution of this term is delicate, particularly the role of the interaction between species fluctuations and chemical reactions and micromixing influence. Moreover, in the  $\beta$ -PDF model described by [9], non reactive species as nitrogen are taken into account for global variance calculation. Adaptation of this model to the PaSR case has been proposed in order to limit the influence of nitrogen fluctuations on combustion calculation. The weak point of the the presumed PDF is the assumption of statistical independence between the temperature and the gas composition proposed in the  $\beta$ -PDF definition (Eqn. 15). Because of this practical simplification from a mathematical point of view, the mean production rate for each species is determined from two independant assumptions on the temperature and the concentrations PDF. In our case of PaSR (adiabatic conditions on walls and stationary case), total enthalpy inside the combustor is constant. Thus, a direct relation exists between sensible enthalpy (mean temperature) and composition described by the enthalpy balance equation (Eqn. 5). As a consequence, temperature fluctuations are functions of the composition fluctuations. In the case of non-premixed combustion, total enthalpy depends on a mixture fraction scalar. In this case, temperature repartition is linked to mixture fraction fluctuations. To conclude, for each case of premixed or non-premixed turbulent combustion, temperature and concentrations PDF are intimately connected through the enthalpy conservation. Then, the link between gas composition and temperature must be taken into account in the assumed-PDF definition. Based on the  $\beta$ -PDF approach, it is impossible to define a joint PDF for temperature and concentrations because of integration of Arrhenius exponential terms which can’t be solved easily using  $\beta$ -PDF mathematical benefits. Thus, if concentrations and temperature fluctuations are totally decorrelated, non-physical solutions are considered for the chemical production rates

calculation and great errors are introduced for the very reactive reactions like hydrogen-air combustion. Nevertheless, in the studied range and for the two fuels, the Vulis models and the IEM model give very close results. Moreover, the Vulis model is simpler than the IEM model and has a low CPU cost.

It is relevant to notice that the critical extinction limit is closely linked to the molecular mixing characteristic time  $\tau_m$ . Actually, when  $\tau_m$  is very small ( $\tau_m < 10^{-5}s$ ), the micromixing is good and combustion is not affected. Then, the solution is quite close to the PSR case. For this configuration, combustion is governed by the chemical kinetics of reactions. If the micromixing time becomes larger, the critical residence time of extinction increases strongly and tends to  $\tau_m$ . In this case, extinction of combustion is governed by the turbulence-chemistry interactions inside the combustor.

Based on the MIT micro turbine geometry, mean velocity is evaluated and, in a first approximation, the characteristic time of molecular mixing is estimated to be 0,1 ms. This value is very close to the characteristic residence time of micro systems previously mentioned. Calculations based on MIT micro turbine operating conditions indicate, in a first approximation, that maximum temperature level reached for a hydrogen-air combustion is near 1800 K, and near 1650 K for a methane-air combustion. Furthermore, we can notice that temperature decreases quickly for residence times smaller than  $10^{-3}s$ . Thus, to guarantee a high intensity combustion and flame stabilization, the mean residence time inside the combustor must be greater than  $10^{-3}s$ , especially for methane-air combustion. Nevertheless, as shown previously, this limit can be improved with a better mixing inside the combustor. Then, 3-D reactive flow calculations will be performed and several geometries of combustion chamber with large recirculation zones will be proposed.

## 5 Conclusions

A parametric study is performed with a Perfectly Stirred Reactor model to determine the sensitivity of combustion limits to significant parameters of micro systems and to different fuels. It appears that heat losses, increased by scale-down, are very significant for combustion efficiency. With realistic values of heat losses, the critical time of extinction limit can rise dangerously and reaches the same order of magnitude as the characteristic residence time of the combustor, especially for hydrocarbon fuels. Those results will be compared with complete CFD calculations and with experimentation results.

Furthermore, in micro volumes, the time required for having a complete mixing at molecular scale can be of the same order of magnitude as the mean residence time. Thus, influence of partial mixing on combustion is particularly studied with three different models of micromixing. We have shown that results given by the  $\beta$ -PDF approach are quite different from the others micromixing models and differences are explained by physical inconsistencies. The cheaper one in CPU cost (Vulis model) will be chosen to be included in a CFD code for global calculations previously mentioned. The micromixing time appears as a crucial parameter and sets the minimum combustor volume for a given mass flow rate. Particle age repartition inside combustor must be improved in order to support exchange between fresh and hot gases, and thus, maintain a stabilized flame. For example, recirculation zones allow to keep burnt gases in a hot region of the combustor and favour the combustion stabilization.

This work is limited to the study of stationary states. It could be interesting to study the time needed to reach high intensity combustion at ignition and the time needed to return to lower temperature when extinction is observed. If the time needed to extinguish combustion is long, the critical limit can be overstepped without an immediate extinction of combustion. A security factor on extinction limit must be respected in order to prevent residence time fluctuations and a strong temperature decrease. In future works, a same parametric studies will be performed with a direct injection of fuel inside the combustor. Results will be compared with the premixed case. Then, several geometries of combustor will be studied experimentally to determine the effect of residence time distribution on mixing



efficiency and combustion stabilization. In parallel, combustion in those geometries will be studied numerically with the modified CFD code.

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