# Experimental Study of the Diluent Influence ($N_2$, He, Ar) On Stable Premixed Methane Flame in a Quartz Micro Flow Reactor

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## 1 Introduction

The current strong interest in the miniaturization of electromechanical systems (MEMs) [1] is related to an increasing demand on portable power generation devices with high-density energy. One solution investigated to address this issue is to exploit the high-density energy of hydrocarbon fuels. The driving idea is that, even with low energy efficiency, devices using the combustion of hydrocarbon fuels will provide more energy than the most powerful current batteries [2]. Power generation devices requiring to be as small as possible, reactors used in micro scale combustion have an inner diameter that is typically smaller than the quenching diameter. At this scale, characteristic flame behaviors have been experimentally [3] and numerically [4] observed such as flames with repetitive extinction and ignition (FREI), weak flames and stable flames depending on the inlet velocity.

Previous studies investigated fuel/air mixtures [5]–[7] but since application to micro scale combustion are numerous, a deep understanding of flame behaviors in different environments is required. Therefore the present work focuses on the influence of different diluent on premixed stable $CH_4$ flame in order to point out physico-chemical properties that have an impact on stable flame in micro-flow reactor.

## 2 Experimental set-up

The experimental set-up used in this study has been described in a previous paper [5] and is shown in Figure 1. A cylindrical quartz micro-flow reactor (1.85 mm inner diameter in this study) is externally heated by

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three $H_2/O_2$ micro blowtorches (Spirig blowtorches). This heating system induce a homogenous temperature distribution around the surface and a temperature gradient along the tube, reducing heat losses and allowing the combustion in diameters smaller than the standard quenching diameter. An infrared camera (FLIR A655sc) continuously monitors the external temperature of the quartz tube. In this study, two different systems were used to determine the flame position in the reactor. Results for $CH_4$/air flames, taken from [5], and $CH_4/O_2/Ar$ used a Princeton Instrument spectroscopy EMCCD camera (ProEm 1600). Results for diluents containing He used a Phantom v1611 camera coupled with a High-Speed IRO intensifier. The use of two different cameras is due to the availability of the instrument and is reputed to have no impact on the results. A 430 nm passband filter is used to detect the CH* emission from the flame.

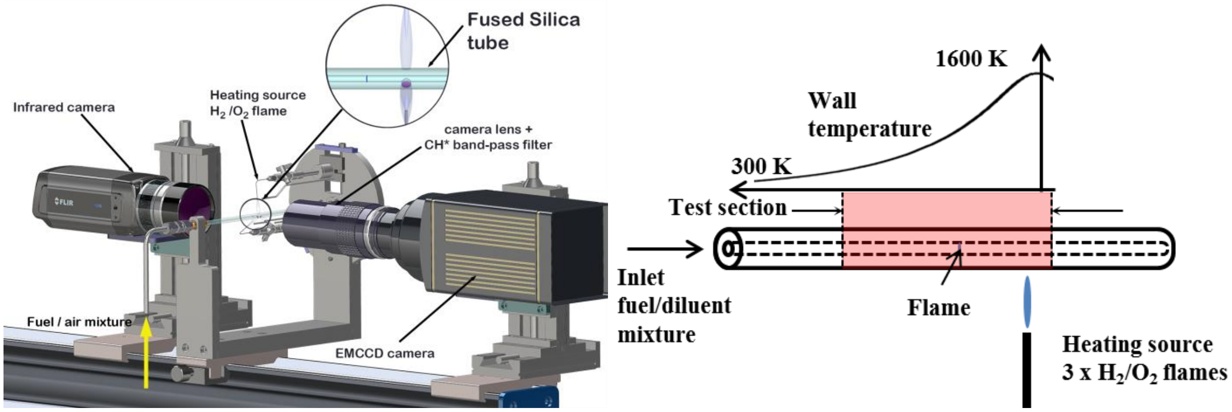

Figure 1. Left: schematic of the experimental set-up; right: schematic of the temperature profile along the channel (test section length of 0.03 m).

## 3 Results

In order to study the impact of diluents on stable flame in micro-flow reactor, all experiments taken into account are for stoichiometric methane premixed flame, in the same tube with an inner diameter of 1.85 mm and with the same temperature gradient applied on the wall. The only parameters varying in this study are diluents used in the premixed flame and inlet velocities. The mole fraction of $O_2$ is kept constant between each diluent in order to avoid chemical effects. Results for stoichiometric $CH_4$/air flames are extracted from previous experiments by Di Stazio et al. [5] whereas new results with others diluents were obtained during for this work and are gathered in Table 1.

Table 1: Compositions of the diluents used in this study in mole fraction

| Diluent | $O_2$ | $N_2$ | He | Ar |
|---|---|---|---|---|
| $N_2$ | 21.0 % | 79.0 % | 0 % | 0 % |
| $N_2$/He | 21.2 % | 39.6 % | 39.2 % | 0 % |
| He | 21.7 % | 0 % | 78.3 % | 0 % |
| Ar | 21.6 % | 0 % | 0 % | 78.4 % |

Regardless of the diluent, the increase of inlet fresh gases velocity implies the displacement toward hotter wall region of the stable flame location as can be observed from Figure 2a). Under these experimental conditions, the variation of this displacement with the increase of the inlet velocity follows a linear trend for each diluent. A linear regression is used to describe the variation of stable flame location with the inlet velocity. In this study, the different slopes observed between these four set of data are assumed to be due to thermodynamics and transport properties of each mixture. In order to investigate these aspects, pure species heat capacities were calculated using a 4th order polynomial in temperature following the NASA chemical equilibrium code formalism [8]. Dynamic viscosities were also calculated with standard kinetic theory [9] and thermal conductivities using the method developed by Peters and Warnatz [10], with the temperature dependence of the rotational relaxation collision number given by Brau and Jonkman [11]. Then, the mixture-averaged thermodynamics and transport properties were calculated. Heat capacities of the different mixtures were deduced from the ideal gas theory, the viscosity with the Wilke's formula [12] and the thermal conductivity following Mathur et al. [13] recommendations.

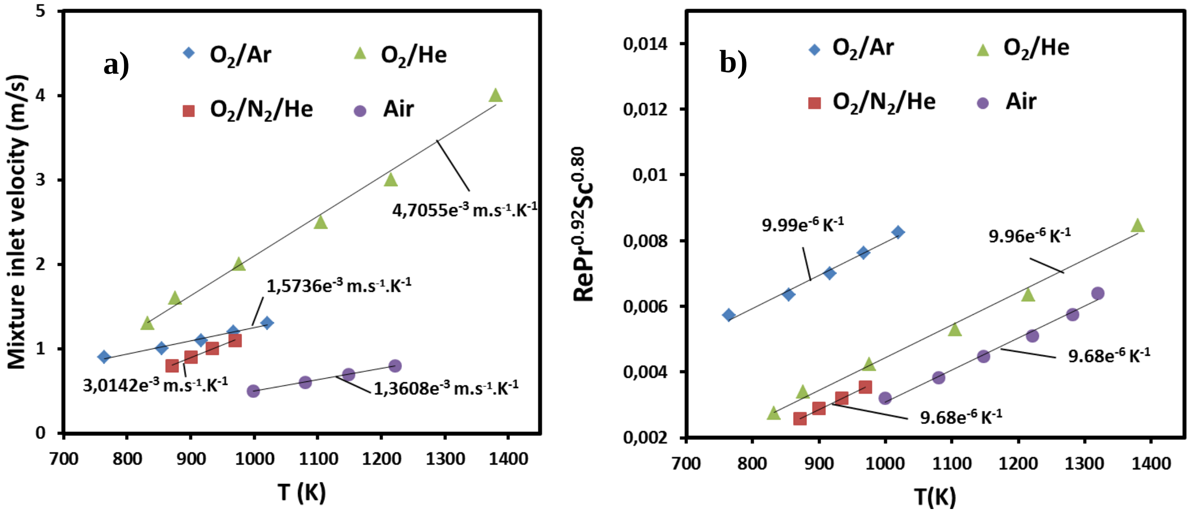

Figure 2: a) Temperature stabilization of stoichiometric $CH_4$ flames as a function of mixture inlet mean velocities for different diluents; b) Temperature stabilization as a function of $RePr^{0.92}Sc^{0.80}$.

In order to point out physical properties playing an important role in slopes observed in the Figure 2 a), we calculate the Reynolds number (Re), the Prandtl number (Pr) and the Schmidt number (Sc) for each mixture and each inlet velocity. These dimensionless numbers are calculated in the fresh mixture, considering a temperature of 300 K. The mass diffusivity of methane in the mixture, calculated with the formula developed by Bird et al. [14], is used in order to compute Sc. These three dimensionless numbers allow us to take into account several properties playing an important role in the thermodynamic behavior of the fluid. As results in Figure 2 a) show a linear dependence between the inlet velocity and the flame location, a combination of these three dimensionless numbers is used as proportionality coefficient. To extract the weight of each coefficient, the results are all assigned with an exponent $Re^xPr^ySc^z$. The coefficient "x" is constraint to a value of 1. Indeed, a linear dependence is assumed between the inlet velocity and the temperature, and because the velocity appears only in Re. A nonlinear general reduced gradient method is used to vary "y" and "z" in order to get the same proportionality coefficient describing the linear relation between the four diluents. In Figure 2 b), slopes between diluents are very similar with a relative difference between the biggest and the smallest slope being less than 3.5 %. This figure shows that, for the four different mixtures tested here, the quantity $RePr^{0.92}Sc^{0.80}$ is proportional to the stable flame location identified with the external wall temperature.

$$Re\,Pr^{0.92}\,Sc^{0.80} = \frac{d\mu^{0.72}C_p^{0.92}\rho^{0.20}}{\lambda^{0.92}D^{0.80}}u_{inlet} = Cu_{inlet} = \mathrm{a}\frac{T}{T_0} + b \qquad (1)$$

In the Eq. (1), "C" and "b" depend on diluents whereas "a" is the slope and is considered to be the same for each diluent in the following part. Eq. (1) shows that the heat capacity ($C_p$) and the thermal conductivity ($\lambda$) of the mixture play a more important role than the dynamic viscosity ($\mu$) or the mass diffusivity of methane in the mixture (D) in the determination of the coefficient "C". As expected, $C_p$, (linked to the adiabatic flame temperature) and $\lambda$ are relevant properties but cannot describe alone stable flames behavior in the micro-flow reactor. The weight of Pr higher than that of Sc meaning that the ratio between viscosity and thermal diffusivity plays a more important role than the ratio between viscosity and mass diffusivity (of methane in the mixture) in the stable flame behavior.

Table 2: Laminar burning velocity, adiabatic flame temperature, transport and thermodynamics properties of a stoichiometric mixture $CH_4/O_2$/diluent (@ 300 K, p=1 atm)

| Diluent | Laminar burning velocity (m $s^{-1}$) | Adiabatic flame temperature (K) | Thermal conductivity ($10^{-2}$ W $m^{-1}$ $K^{-1}$) | Specific heat capacity (J $kg^{-1}$ $K^{-1}$) | $CH_4$ mass diffusivity ($10^{-5}$ $m^2$ $s^{-1}$) |
|---|---|---|---|---|---|
| $N_2$ | 0.3 | 2221 | 2.65 | 1075 | 2.34 |
| $N_2$/He | 0.61 | 2367 | 5.48 | 1401 | 3.11 |
| He | 1.19 | 2541 | 9.08 | 2240 | 4.54 |
| Ar | 0.77 | 2537 | 2.03 | 661 | 2.32 |

The addition of He in the diluent strongly increases the thermal conductivity of the mixture as can be seen in Table 2. The high thermal conductivity coupled with a high adiabatic flame temperature (case $O_2$/He) leads to strong heat losses to the wall. With Ar as diluent, the thermal conductivity of the mixture is the lowest while the adiabatic flame temperature is similar to that of $O_2$/He mixtures which leads to lower heat losses to the wall. Thus, fresh gases are more preheated with He than with Ar, allowing the flame to stabilize at lower temperature. Moreover, the time between the ignition of the mixture and the stabilization of the flame is also longer with Ar than with He due to thermal equilibrium longer to achieve. The strong influence of the thermal conductivity and the specific heat capacity in equation (1) confirms the importance of flame wall thermal interaction on the combustion in micro-flow reactors.

Equation (1) can be used to determine the stable flame location sensitivity to the inlet velocity for a given mixture. If the sensitivity to the inlet velocity is too high, meaning that the slope in figure 2) a) is too small, it indicates a flame difficult to stabilize with a higher risk of flashback.

## 4 Conclusions

Experiments were performed with premixed stoichiometric methane flame with four diluents. It was observed that stable flames temperature locations linearly depends on inlet velocity with proportional coefficients varying with the diluent. These coefficients were linked with mixture thermodynamic and transport properties in order to sum up four different behaviors in one single relation. This present work underlines the fact that there are no simple relations between laminar flame speed, adiabatic flame temperature and stable flame position in a micro-flow reactor. The role of transport properties needs to be further investigated either experimentally or numerically.

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