# Pulsating combustion of ethylene in micro-channels with controlled temperature gradient

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# **1** Introduction

Micro combustion is now a very topical issue, and despite the considerable progress made in the last decade [1, 2], many aspects are still unknown. Currently, different fuels have been tested experimentally to study their ignition characteristics in micro-channels [3-5]. However, there is still a lack of clearly defined information about the dynamics of stable and unstable flames at micro-scale. As illustrated in our previous paper [6], our experimental device allows to characterize the frequencies of unstable flames under various experimental conditions while ensuring a uniform temperature distribution. Moreover, the presence of oscillating flames, already observed experimentally [6, 7] and numerically [8-11], has been confirmed under specific conditions in the transitions between the different flame regimes, but the knowledge on these instabilities is still very limited.

Ethylene is one of the main intermediate in the combustion of real fuels, and thus plays a critical role in governing the kinetics and the heat release. As such, understanding its combustion behavior under micro combustion regime is crucial for the development of this combustion mode. Kikui et al. [12] studied ethylene combustion in the weak flame regime and showed with the help of numerical simulations that it is oxidized into formyl radicals either directly or through the formation of formaldehyde CH<sub>2</sub>O. Furthermore, the large production of CH\* radicals during ethylene combustion makes this fuel suitable for the experimental device used here.

The purpose of this work is to investigate the combustion characteristics of this key intermediate under different flame regimes (stable flames, FREI, and weak flames) as a function of the equivalence ratio. For unstable flames, the flame dynamics was characterized with a high-speed camera. This equipment allowed visualizing for the first time the spatial progression of unstable flames at high frequency. Also, the oscillating behaviors and the flame obtained under very lean conditions are discussed.

### 2 Experimental set-up

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A schematic of the experimental setup is shown in Fig. 1 (left). C<sub>2</sub>H<sub>4</sub>/air mixtures were supplied in a quartz tube with an inner diameter of 1 mm at atmospheric pressure. A smaller diameter than for methane was here necessary since ethylene has a smaller quenching diameter [13]. The channel was heated externally by three  $H_2/O_2$  flames (Spirig blowtorches) to ensure a stationary temperature profile from ambient temperature to about 1600 K (Fig. 1, right). The temperature along the outer side of the channel was continuously measured by a FLIR A655sc thermal camera. An emissivity correction was carried out, since the emissivity of the fused silica depends on the wavelength and the temperature [14]. In order to confirm the accuracy of our measurements, the temperature profile along the inner surface of the tube wall was also measured with a inconel-sheated K-type thermocouple (diameter 0.25 mm). A Princeton Instrument spectroscopy EMCCD camera, equipped with an optical band-pass filter (20BPF10-430), was used to collect the CH\* chemiluminescence indicating the flame position. The acquisition frequency was set at 4260 Hz and pixel binning was performed. The signal was amplified (high EM gain) and the spatial resolution was 62 pixels/mm. In order to track the unstable flames propagation, a high-speed camera Phantom V1611, equipped with a teleconverter Teleplus MC7, was employed. A sample rate of 12000 fps and an exposure time of 82 us were adopted to simultaneously optimize picture brightness and temporal resolution.



Figure 1. Left: Schematic of the experimental device; Right: Temperature profile along the channel.

#### **3** Results

As observed previously [6, 15] for methane/air mixtures, three flame regimes exist depending on the inlet velocity: stable flames at high speed (> 0.7 m s<sup>-1</sup>), FREI (Flames with Repetitive Extinction and Ignition) for lower speeds, and weak flames at very low speed (< 0.2 m s<sup>-1</sup>). In some conditions, oscillatory flames were observed between the different regimes: oscillating FREI in the transition between stable flames and FREI, and oscillating weak flames in the transition between FREI and weak flames. Results obtained over an extended range of equivalence ratios (from 0.3 to 1.5) are discussed below. Peculiar flame behaviors were observed under very lean conditions ( $\phi = 0.3$ ).

Figure 2 shows the results obtained when varying the equivalence ratio. As observed previously for methane/air mixtures [6], the stabilization temperature of the stable flames decreases with the inlet velocity. Norton and Vlachos [10] explained that when the inlet velocity is increased it requires a longer distance to preheat the fresh gases: the flame is therefore stabilized further downstream. The wall temperature at the flame location is hence higher and the heat losses are lower, thus resulting in higher reaction rates and higher flame temperatures. In the FREI regime, ignition occurs at high temperature (> 1100 K), and then the flame propagates upstream until extinction. Weak flames were observed at equivalence ratios of 1.0 and 1.5 and oscillating weak flames were observed at  $\varphi = 0.9$  and 1.0. At  $\varphi = 0.4$ , oscillating flames were detected between the stable flames and FREI.

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By comparing the results obtained for different equivalence ratios, one can notice that the minimum stabilization temperatures are obtained for  $\varphi = 1.1$  and 1.3. These equivalence ratios correspond to ethylene/air mixtures for which the laminar flame speed is maximum.



Figure 2. Flame position as a function of inlet velocity and wall temperature as a function of the equivalence ratio: 0.3 (grey), 0.4 (blue), 0.7 (orange), 0.9 (red), 1 (black), 1.1 (green), 1.3 (purple), 1.5 (pink).



Figure 3. Flame images and corresponding CH\* signals at  $v = 0.6 \text{ m s}^{-1}$  and  $\phi = 1$ : a) ignition, b) propagation, c) time before extinction.

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Regarding FREI, the experiments performed with the high-speed camera allowed tracking the flame inside the channel at high frequency. Figure 3 depicts the three main phases of the flame progression: ignition, propagation and time before extinction. The images obtained (top) are compared to the CH\*signals recorded with the EMCCD camera (bottom).

The flame ignition (a) is characterized by a low luminosity. When it propagates (b), the brightness and the thickness of the flame increase as illustrated by the CH\* profile: the signal is 4 mm thick. Before extinction (c), the flame gets thinner with a concave shape and the CH\* profile becomes sharper with a higher intensity. The thickening of the flame front during the propagation phase cannot be attributed to an accumulative phenomenon. Experiments were carried also at 20000 fps and similar flame thicknesses were observed. Images at 12000 fps were chosen in order to get higher luminosity. As compared to stable flames, one of the interests of this FREI region is that the ignition phenomenon can be observed. As can be seen from Fig. 2, fuel-lean mixtures ignite at significantly lower temperatures than stoichiometric or fuel-rich mixtures: for instance, at v = 0.4 m s<sup>-1</sup>, the mixture at  $\varphi = 0.4$  ignites at T = 1168K whereas the stoichiometric mixture ignites at T = 1210K.

The flames observed under very lean conditions ( $\varphi = 0.3$ ) and high inlet velocity have a very low brightness, which is commonly associated with weak flames. These weak flames stabilize in the high temperature region of the temperature gradient. Decreasing the inlet velocity (down to  $v = 0.45 \text{ m s}^{-1}$ ) results in a flame stabilization at lower and lower temperatures. Then for inlet velocities below 0.45 m s<sup>-1</sup>, oscillating weak flames were observed (Fig. 2). Figure 4 (left) shows the frequency of these oscillating flames obtained through a Fourier Transform analysis of the spectral cross-section of the CH\* signal. The results show that the frequency is very high but slightly decreases with the inlet velocity. The time evolution of the spatially integrated CH\* signal is displayed in Fig. 4 (right). One can notice that the luminous intensity varies randomly, while for the oscillating flames observed between the stable flame and FREI regimes, the luminosity fluctuates regularly as will be detailed in Figure 5. The spatial displacement of these oscillating weak flames is of about 0.5 mm, which corresponds to a wall temperature difference of 40 K. These unstable flames are similar to FREI. Indeed, the heat released by the reaction decreases with the inlet velocity, leading to a near blow-off condition [10]. The flame does not propagate, but it is "trapped" in the high temperature zone. During the pulsations, the radical concentration fluctuates, whereas the radical pool is totally depleted before the re-ignition for a typical FREI, as explained by Miroshnichenko et al. [11].



Figure 4. Left: Frequencies at  $\varphi = 0.3$ ; Right: Temporal evolution of CH\* signal at v = 0.45 m s<sup>-1</sup>.

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At an equivalence ratio of 0.4, oscillating flames were observed in the transition region between stable flames and FREI (Fig. 5), between v = 0.53 and v = 0.48 m s<sup>-1</sup>. These flames oscillate at high frequency but do not extinguish, i. e. the CH\*signal does not go to zero. The oscillation amplitude increases when decreasing the inlet velocity until getting a typical FREI at v = 0.47 m s<sup>-1</sup>. The mathematical model proposed by Jackson et al. [9] was able to predict these instabilities that are due to the heat losses through the tube wall. We have also observed oscillating weak flames at equivalence ratios of 0.9 and 1.0, but in the region between the FREI and the weak flame regimes.



Figure 5. Oscillating flame observed at  $v = 0.5 \text{ m s}^{-1}$  and  $\varphi = 0.4$ .

Figure 6 shows the evolution of the frequencies obtained under different conditions. The lowest frequencies were recorded  $\varphi = 0.9$  and 1.3. The oscillating frequencies increase as the equivalence ratio varies towards fuel-rich or fuel-lean conditions. The frequency peaks for the oscillating flames. The results were compared to those obtained for a stoichiometric methane/air mixture, obtained with a 1 mm inner diameter tube. Even in this case, we observed high frequency oscillating flames in the transition between stable flames and FREI. The frequencies are comparable to those obtained with ethylene/air mixtures under fuel-lean conditions.



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Figure 6. Flame frequencies obtained varying the inlet velocity at different equivalence ratios.

# 4 Conclusions

Our experimental set-up confirms its interest in the study of small-scale combustion. Experiments were performed with ethylene/air mixtures in a 1 mm inner diameter tube over a wide range of equivalence ratios. Under very lean condition the combustion dynamics changed: at an equivalence ratio of 0.3, only weak flame and oscillating weak flame were observed. Furthermore, oscillating flames were observed at equivalence ratio of 0.4 between the stable flames and the FREI region. For mixtures with equivalence ratio between 1.1 and 1.3, the flames stabilize at lower wall temperatures. Furthermore, it was possible to carry out a frequency analysis, demonstrating that the highest frequencies are recorded for oscillating flames.

# References

[1] Ju Y, Maruta K (2011). Microscale combustion: Technology development and fundamental research. Prog. Energy Combust. Sci., 37 (6): 669.

[2] Kaisare NS, Vlachos DG (2012). A review on microcombustion: Fundamentals, devices and applications. Prog. Energy Combust. Sci., 38 (3): 321.

[3] Kamada T, Nakamura H, Tezuka T, Hasegawa S, Maruta K (2014). Study on combustion and ignition characteristics of natural gas components in a micro flow reactor with a controlled temperature profile. Combust. Flame, 161 (1): 37.

[4] Nakamura H, Tanimoto R, Tezuka T, Hasegawa S, Maruta K (2014). Soot formation characteristics and PAH formation process in a micro flow reactor with a controlled temperature profile. Combust. Flame, 161 (2): 582.

[5] Hori M, Nakamura H, Tezuka T, Hasegawa S, Maruta K (2013). Characteristics of n-heptane and toluene weak flames in a micro flow reactor with a controlled temperature profile. Proc. Combust. Inst., 34 (2): 3419.

[6] Di Stazio A, Chauveau C, Dayma G, Dagaut P (2016). Combustion in micro-channels with a controlled temperature gradient. Exp. Thermal Fluid Sci., 73: 79.

[7] Tsuboi Y, Yokomori T, Maruta K (2008). Study on ignition and weak flame in heated meso-scale channel. In: ASME International Mechanical Engineering Congress and Exposition, Proc, 6: 155.

[8] Minaev S, Maruta K, Fursenko R (2007). Nonlinear dynamics of flame in a narrow channel with a temperature gradient. Combust. Theory Modelling, 11 (2): 187.

[9] Jackson TL, Buckmaster J, Lu Z, Kyritsis DC, Massa L (2007). Flames in narrow circular tubes. Proc. Combust. Inst., 31 (1): 955.

[10] Norton DG, Vlachos DG (2003). Combustion characteristics and flame stability at the microscale: a CFD study of premixed methane/air mixtures. Chem. Eng. Sci., 58 (21): 4871.

[11] Miroshnichenko T, Gubernov V, Minaev S, Maruta K (2015). Diffusive-thermal instabilities of high Lewis number flames in micro flow reactor. In: Radulescu MI (Ed.) 25<sup>th</sup> International Colloqium on the Dynamics of Explosions and Reactive Systems, Leeds, UK, 2015.

[12] Kikui S, Nakamura H, Tezuka T, Hasegawa S, Maruta K (2015). Study on combustion and ignition characteristics of ethylene, propylene, 1-butene and 1-pentene in a micro flow reactor with a controlled temperature profile. Combust. and Flame, 163: 209.

[13] Gutkowski A (2006). Laminar burning velocity under quenching conditions for propane-air and ethylene-air flames. Arch. Combust., 26 (3/4): 163.

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[14] Sova RM, Linevsky MJ, Thomas ME, Mark FF (1998). High-temperature infrared properties of sapphire, alon, fused-silica, yttria, and spinel. Infrared Phys. & Tech., 39 (4): 251.
[15] Maruta K, Kataoka T, Kim NI, Minaev S, Fursenko R (2005). Characteristics of combustion in a narrow channel with a temperature gradient. Proc. Combust. Inst., 30 (2): 2429.