

Study of soot aggregate formation and oxidation in swirled stratified premixed ethylene/air flames

Andrei-Silviu Milea¹, Aurélien Perrier^{1,2}, Marcos Caceres¹,
Gilles Godard¹, Alexis Vandael¹, Gilles Cabot¹, Frédéric Grisch^{1,*}
1: Normandie Univ., UNIROUEN, INSA Rouen, CNRS, CORIA, 76000 Rouen, France
2: Safran Aircraft Engines Villaroche, 77550 Moissy-Cramayel, France
* Correspondent author: frederic.grisch@coria.fr

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ABSTRACT

Soot particles are one of the main causes of today's pollution because of their negative contribution to global warming and human health. Aviation is one of the domains dealing with soot reduction as the new engine concepts are developed to reduce fuel consumption and global emissions. Despite the fact that most combustion devices used for air transportation operate at high pressure (e.g., aircraft gas turbines up to 40 bar), our understanding of soot formation and oxidation in such conditions is not yet at an appropriate level, as there is still a fundamental lack of experimental data and corresponding predictive models in the literature. Thus, the objective of the current study is to evaluate soot formation and oxidation processes in stratified, swirled, premixed ethylene/air flames examined with a variety of laser diagnostics designed to simultaneously measure soot particle and soot precursor 2D-distributions, as well as the flame structure and the aerodynamic field. For that, the SIRIUS burner was selected because of its ability to produce flames with topologies similar to those encountered in aircraft combustors. Soot particle distributions are measured by Planar Laser-induced incandescence (PLII) diagnostic. The flame structure is obtained by detecting the hydroxyl radicals (OH) with Planar laser-induced fluorescence (PLIF). A second PLIF diagnostic is also used to investigate the production of polycyclic aromatic hydrocarbons (PAH) with the detection of two benzene rings molecules which are recognized as good precursors of soot nucleation and growth. Finally, the particle Image Velocimetry (PIV) diagnostic is used for measuring the velocity distributions. These laser diagnostics are coupled together in order to obtain cross-correlations between several scalar parameters playing a determining role in the soot formation/consumption processes. The experimental results collected at atmospheric pressure are reviewed and critically assessed. A scenario describing the link between the soot inception, growth, aggregation and oxidation processes is proposed by analyzing velocity, OH, PAHs and soot distributions. In particular, the data reveal the presence of distinct regions for these processes. Incipient soot production zone is strongly function of specific local conditions of velocity, PAH concentration, and strain rate encountered at the interface of the internal recirculation zone and the fuel/air jet. The central part of the inner recirculation zone in which large structures move at low velocities provides suitable conditions for the aggregation of nascent soot particles while an oxidation region located in the upper zone of the internal recirculation zone favors the consumption of soot.

1. Introduction

The combustion of conventional fossil fuels leads to the production of smoke which usually contains heavy particles of soot (Houghton et al., 2001; Penner et al., 2003). The presence of these microscopic airborne particles in the atmosphere not only enhances the global warming effect but affects human health as it creates deadly respiratory diseases as well (Niranjan & Thakur, 2017). One of the domains where this challenge needs to be addressed is air transportation. Pollutants produced from aircraft engines pose serious public health and environmental issue (Ramanathan & Carmichael, 2008). Furthermore, international aviation emissions could be multiplied by a factor 3 by 2050 (ICAO, 2016). Although the global pandemic of COVID-19 stunted global exchanges in 2020, air traffic is expected to perform a fast recovery and therefore, beat further records in the future years (IATA, 2022). On the other hand, aeronautical emissions are subject to more stringent regulations, amongst others, on greenhouse gases (i.e. CO₂), and pollutants such as nitrogen oxides (NO_x) and particulate matter (Valdés et al., 2019). To tackle this problem, aircraft motorists need to develop ever more innovative engine architectures, able to reduce emissions and fuel consumption, while maintaining high combustion efficiency. Among different strategies investigated, new injection systems based on multi-point and Rich-burn, Quick-quench, Lean-burn concepts have been identified by aircraft motorists as promising technologies in terms of reduction of NO_x-emissions. However, this one presents a high risk of particulate emissions due to the presence of the rich flame zone intended to ensure flame stability. In that context, a fine comprehension of the soot inception, growth, and oxidation is required. Despite many efforts taken over the last decades, the full understanding of processes previously mentioned remains a challenging task, namely due to the many parameters and phenomena involved.

In order to study the formation and oxidation of soot in aeronautical flames, a stratified premixed swirl burner is used to reproduce their operating conditions. Called SIRIUS (*Swirl stratified burner for the study of Soot production*), this simplified geometry burner was developed with an ultimate objective of analyzing soot under elevated pressure conditions. The current study presents preliminary results from the simultaneous use of four laser diagnostics on stratified and swirled C₂H₄/air premixed flames operating at atmospheric pressure. This is accomplished by simultaneous imaging the velocity, soot volume fraction as well as OH and PAH distributions using particle image velocimetry (PIV), planar laser-induced fluorescence (PLIF), and planar laser-induced fluorescence (PLIF) of OH and PAHs, respectively. Results collected at 1 bar are examined and critically evaluated with the goal of providing an in-depth understanding of the physical processes governing soot formation and oxidation in these flames. Relationships between soot,

OH and PAH distributions as well as aerodynamics, highlighting combined effects of the flame reactivity, PAH distributions, soot formation intermittency and aerodynamics are presented and discussed.

2. Experimental Apparatus

2.1 Burner configuration

Experiments were performed on the SIRIUS burner designed at the CORIA French laboratory. It was conceived with the aim of reproducing the specific topology of aeronautical Low-NO_x flames allowing a detailed investigation of the various physical and chemical mechanisms that govern turbulent combustion in jet engines. The SIRIUS burner presents an architecture similar to the Cambridge-Sandia Swirl burner (Sweeney, Hochgreb, Dunn, & Barlow, 2012) for which stratified swirled premixed ethylene/air flames can be produced in elevated pressure conditions.

An atmospheric version of the SIRIUS burner proved initially its industrial relevance by the results on the soot production and oxidation obtained through various studies performed by Bouvier (Bouvier et al., 2019). Even though the fuel injection is fully gaseous and the combustion conditions were atmospheric, flames produced by this burner respected the same pattern as a real jet engine combustion. A solid innovation was recently brought to SIRIUS in order to study high-pressure flames. Inspired from the high-performance combustion chamber HERON (High prEssuRe facility for aerO-eN-gine combustion), the novel version of the SIRIUS burner shown in Figure 1 is optically accessible, making possible the study of a single sector of a combustion chamber (Grisch et al., 2016). A high-pressure visualization module with large optical accesses (100 mm x 80 mm) is mounted on the head of the burner to allow the application of various laser diagnostics developed at CORIA (Fig. 1). The visualization module is thermally and mechanically resistant by a water cooling and allow the production of flames up to 5 bar.

The head of the burner is composed of three concentric tubes of 10, 20 and 29.5 mm respectively as displayed in Fig. 2. The central one is sealed with a ceramic cap at its upper end, serving as a flame holder for generating an inner recirculation zone. The annular sections formed by the concentric tubes around the ceramic cap are used to inject ethylene/air premixed jets which feed the flame. Similar to aeronautical architectures, the inner flow (yellow on Figure 1) is purely axial while the outer flow (blue on Figure 1) is swirled. The latter is composed of two air injections: an axial injected flow $\dot{m}_{o,axial}$ oriented upwards and a tangential flow $\dot{m}_{o,tang}$. A stabilizing air coflow is injected through a square porous media placed around the tubes.

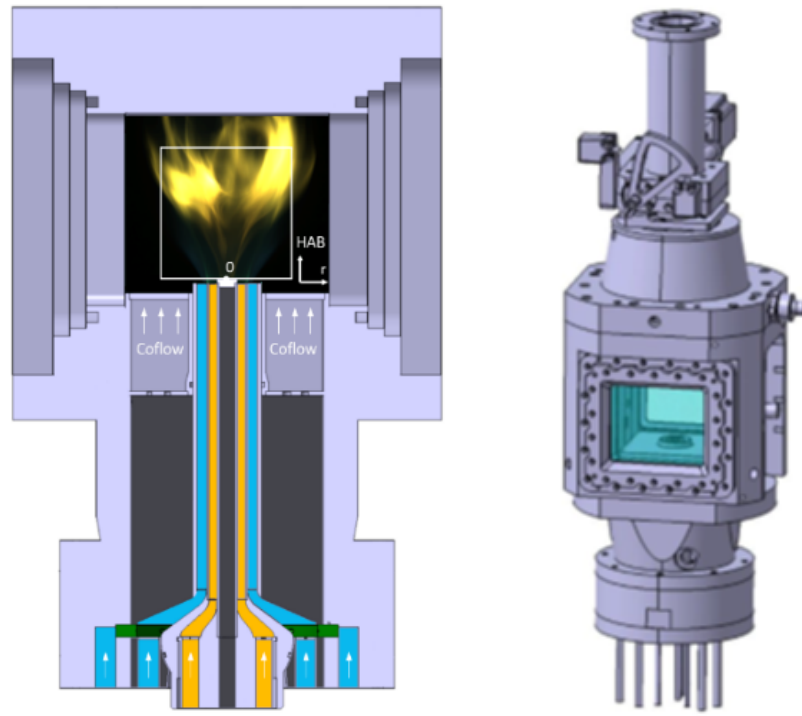


Fig. 1 Global sketch of the SIRIUS burner

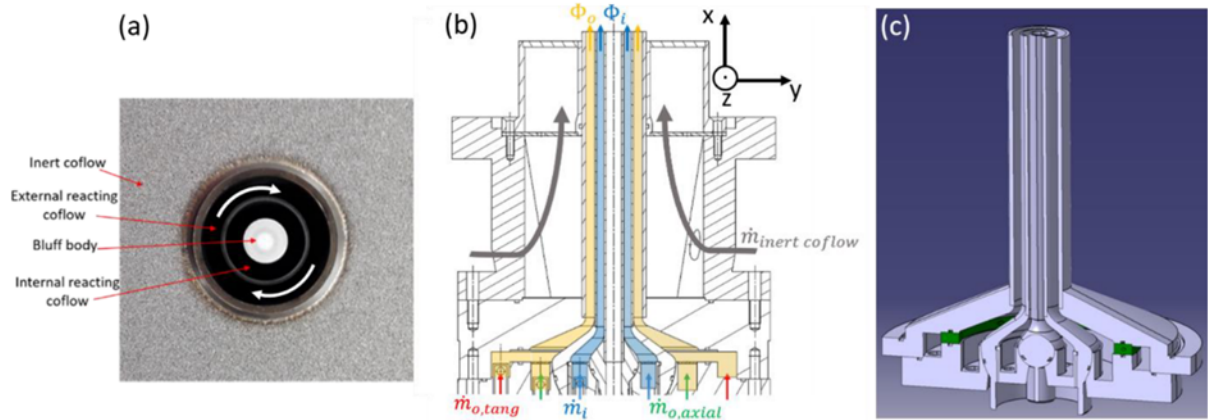


Fig. 2 Detailed view of the fuel injection setup of the SIRIUS burner

In order to limit the analysis of the flames with numerous specific scalar parameters, two main dimensionless parameters are defined to describe the flame operating conditions of the SIRIUS burner which are representative for aerodynamics and chemistry. On the one hand, the swirl number of the flame is characterized by Swirl Flow Ratio (SFR), defined as the ratio between axial and tangential outer flow rates:

$$SFR = \frac{\dot{m}_{o,tang}}{\dot{m}_{o,tang} + \dot{m}_{o,axial}} \quad (1)$$

On the other hand, the equivalence ratios of inner and outer flows are controlled thanks to independent mass flowmeters. The stratification ratio is defined as the ratio between inner Φ_i and outer Φ_o equivalence ratios:

$$SR = \frac{\Phi_i}{\Phi_o} \quad (2)$$

The current study is performed at 1 bar. The inner jet has an equivalence ratio of 2.21 while the outer one equals to 0.74. This corresponds to a stratification ratio of 3. The swirl flow ratio is fixed at 0.25.

	Outer axial		Outer radial		Inner		Coflow
P bar	Air g/s	Fuel g/s	Air g/s	Fuel g/s	Air g/s	Fuel g/s	Air g/s
1	3	0.15	1	0.05	0.6	0.09	6

Table 1 Experimental conditions

2.2 Optical diagnostics

A set of four laser diagnostics was used: OH-PLIF (planar laser-induced fluorescence) to image the flame structure, PLII (planar laser-induced incandescence) to visualize soot particles distributions, PAH-PLIF to observe the zone of soot precursors production, and PIV to measure the aerodynamics.

The laser source for OH-PLIF, PAH-PLIF and PLII diagnostics was a dye laser circulating Rhodamine 6G dissolved in ethanol, pumped by a Nd: YAG operating at 532 nm and generating 6 ns pulses at a frequency rate of 10 Hz. On the other hand, the laser source used for PIV measurements was a double-pulsed 532 nm Nd: YAG laser operating at 10 Hz, and producing a pair of laser pulses with an inter-pulse time of 10 μ s. The arrangement of the laser sources with the burner is displayed in Fig. 3. PLIF and PIV laser beams at the laser exits were converted into 50 mm large, 200 μ m thick laser sheets using a set of optical lenses: a spherical convergent lens ($f=1000$ mm) was used to further focus the laser beam at the probe volume while a combination of a convergent ($f=300$ mm) and a divergent ($f=-25$ mm) cylindrical lenses are used to form a collimated laser sheet. The direction of the laser sheets was adjusted to superimpose them above the burner exit (Fig. 3). The PLIF, LII and PIV measurements were temporally synchronized by positioning the PLIF laser pulse between the PIV laser pulses. The detection of the different signals is provided by a set of four cameras. Three 16-bit emICCD cameras (Princeton Instruments, 1024×1024 pixels) were used for imaging the PLIF and PLII signals, whereas the Mie scattering emitted by particle

seeded into the flow and required for PIV measurements was collected by a double-frame CCD camera (Jai TM4200CL, 2048 × 2048 pixels). A fifth ICCD camera is used to record the flame emission.

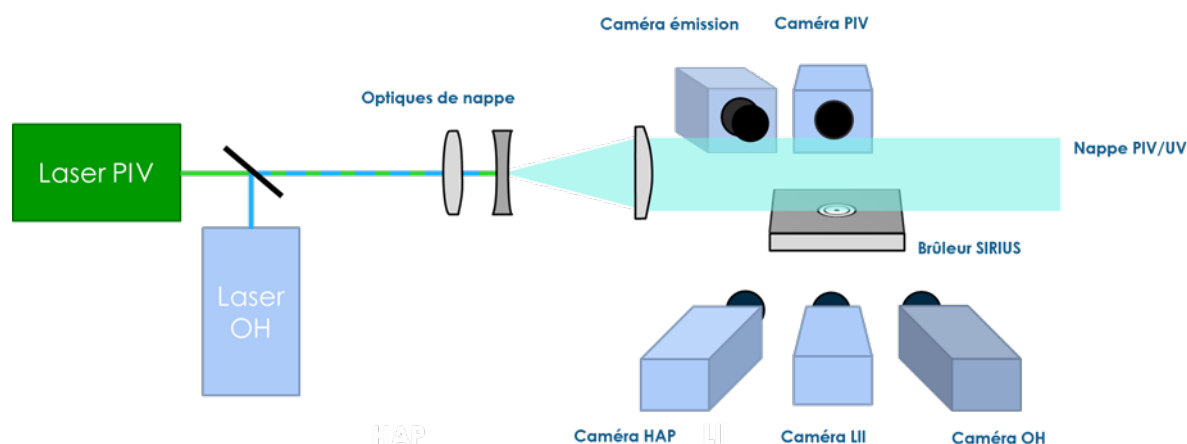


Fig. 3 Schematic of the detection system with both laser sheets of 50 mm large formed by a set of three optical lenses.

2.2.1 PLIF

For the excitation of OH, soot precursors and soot particles, the dye laser is tuned to 282.75 nm, allowing the simultaneous excitation of the $Q_1(5)$ rotational line of the OH electronic band ($A^2X^+(0,0) \leftarrow X^2\Pi(0,1)$), as well as those of naphthalenes (PAH) and soot particles. The $Q_1(5)$ rotational transition was selected for its high-Intensity fluorescence response, its well-isolated spectral feature, and its low dependence on temperature (Kostka et al., 2009). As for the PAH, this wavelength enables the excitation of the mono- and di-aromatic components issued from the thermal decomposition of the fuel. The fluorescence signals of OH radicals and PAHs were recorded with two intensified CCD cameras equipped with UV objectives (CERCO 100 mm f/2.8) and located on one lateral side of the burner (Fig. 3). They were slightly tilted, and equipped with a Scheimpflug mount to correct the focus over the region of study. The OH fluorescence signal was filtered with a combination of a 295 nm long-pass filter and a 315 nm band-pass filter (FWHM = 15 nm). The PAH camera was equipped with a 350-400 nm band-pass filter and a 400 nm short-pass filter.

2.2.2 LII

Soot was detected by the incandescence signals emitted by the particles after their excitation by the laser sheet. The laser beam energy in the laser sheet is fixed around 15mJ per pulse to obtain LII signals with a good SNR. The third intensified CCD camera used for LII was located on the

same side as the PLIF cameras, this one being now perpendicular to the laser sheet (Fig. 3). It was equipped with a Nikon UV-NIKKOR 105mm f/4.5 objective as well as a 400 nm long-pass filter which allowed a strong reduction of the chemiluminescence signal emitted by the flame.

2.2.3 PIV

The aerodynamic field is studied by illuminating particles seeding into the reactive flow with both 532nm laser sheets. ZrO₂ particles of few μm of diameter are selected due to their temperature resistance and their suitable diameter for the reactive flow conditions investigated. The particle motion was recorded by a double-frame CCD camera (Jai TM4200CL, 2048 $\times$ 2048 pixels) placed across the SIRIUS burner, perpendicular to the laser sheet (Fig. 3). The signal was filtered with a 532 nm band-pass filter (FWHM = 5 nm). PIV vectors were computed by cross-correlation of the PIV images using the Davis Software, with a final interrogation window of 16 $\times$ 16 pixels, yielding a 455 μm spacing.

3. Results and discussion

In this section, the images recorded from the laser diagnostics and their correlation are analyzed to obtain a better understanding of the physical and chemical phenomena governing the soot formation at 1 bar. Examples of PLII, PLIF-OH and PLIF-PAH raw signals are displayed in Fig. 4.

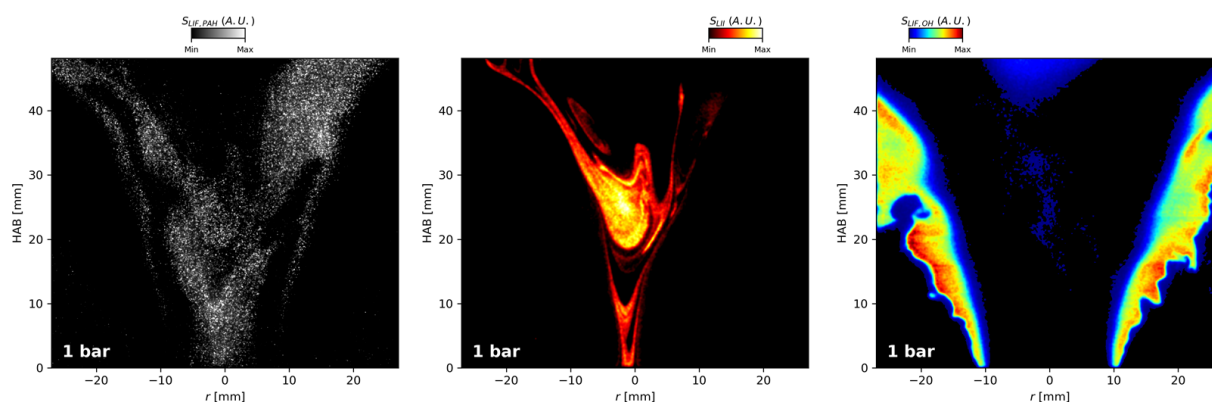


Fig. 4 Raw single-shot 2D distributions of PAH, soot and OH

3.1 Global flow field

In Fig. 5, the superposition of the flame structure with the velocity fields are shown. The left-hand sides show instantaneous images while the right side displays the corresponding time-averaged distributions. The global flame topology (OH-PLIF images) observed in Fig. 4 corresponds to conventional swirl stabilized flames, featuring outer recirculation zones (ORZ) and a bluff-body

used to stabilize the inner recirculation zone along the burner axis (IRZ). As expected, the penetration of the burnt gases arising from the flame zone and going to the burner base is observed. The IRZ and ORZ are separated by the two annular reactive jet flows. Because the flame is confined inside the combustion module, the air co-flow coming from the porous burner initially dedicated to produce a thermal barrier in front of the optical windows, also acts as an outer shield around the flame, closing its sides and stabilizing its shape. Velocities measured in the IRZ (~ 1 m/s) are slightly lower than values from the fuel/air jets (5 m/s for the inner flow and 15 m/s for the outer one). These high velocity differences lead to the production of an annular inner shear layer (ISL) such as and outer shear layer (OSL), underlying large flow fluctuations as well as coherent structures.

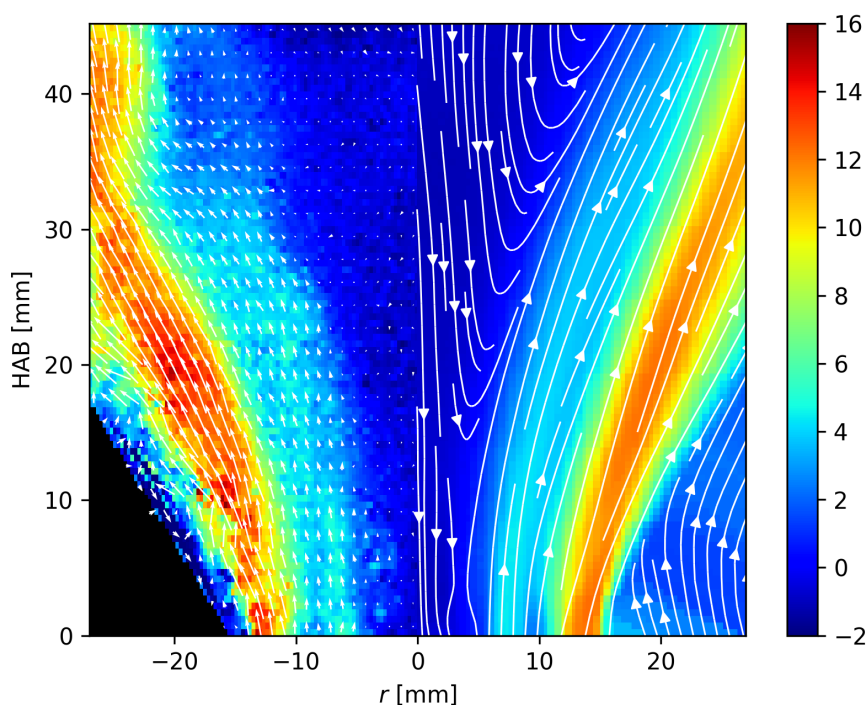


Fig. 5 Results of PIV measurements colorized by vertical velocities. Left: vector field of instantaneous image. Right: mean streamlines

3.2 Flame structure and soot precursors

The hydroxyl radical (OH) is one of the most representative sub-products of combustion reactions. It is created during the decomposition of the air and fuel and subsequently, it gets progressively consumed in the flame due to its reactive nature. Peaks of OH fluorescence are localized within highly reactive zones where combustion reactions are gradually undergoing. These regions called flame fronts, are only a few hundred microns thick and lead to intense heat releases due to the chemical exothermic chemical reactions. Hence, traditionally, the fluorescence signal of the OH

radical can be interpreted to localize flame fronts. Fig. 6 highlights the superposition of the OH-fluorescence signals (jet colors) with the PAH-fluorescence (grey nuances). An analysis of the instantaneous images reveals a high thin intensity gradient of OH within the outer fuel-lean jet, denoting the existence of flame fronts. The signal gradient is highly wrinkled, revealing the high level of turbulence resulting not only from interactions between the outer fuel-lean jet and the external co-flow, but also from intense gas expansion caused by large heat release. The flame is stabilized on the tip of the pipe splitting the inner and the outer fresh jets. It was also observed that OH fluorescence signals don't disappear immediately downstream of the flame front meaning there is a consistent indication of high temperature burnt gas. Weaker OH pockets are also observed into the center of IRZ, suggesting a transport of OH radicals toward the center of the flame due to the aspiration of burnt gases in this area.

On the other hand, PAH signals form a long hollow strip coming from the inner fresh fuel/air jet (Fig. 6). Aromatic species formed in this area result from the thermal decomposition by pyrolysis of the inner fuel-rich jet. The hot gases circulating into the inner recirculation zone heat the ethylene fuel in the vicinity of the injection zone making pyrolysis a dominant phenomenon in this region. On the other hand, no PAH fluorescence signals are visible along the outer fuel-lean jet (0.74 vs. 2.21 for the inner one). Combustion takes place rapidly, creating intense temperature gradients over an area of few millimeters and accelerating the consumption of fuel and its thermal decomposition products by exothermic chemical reactions.

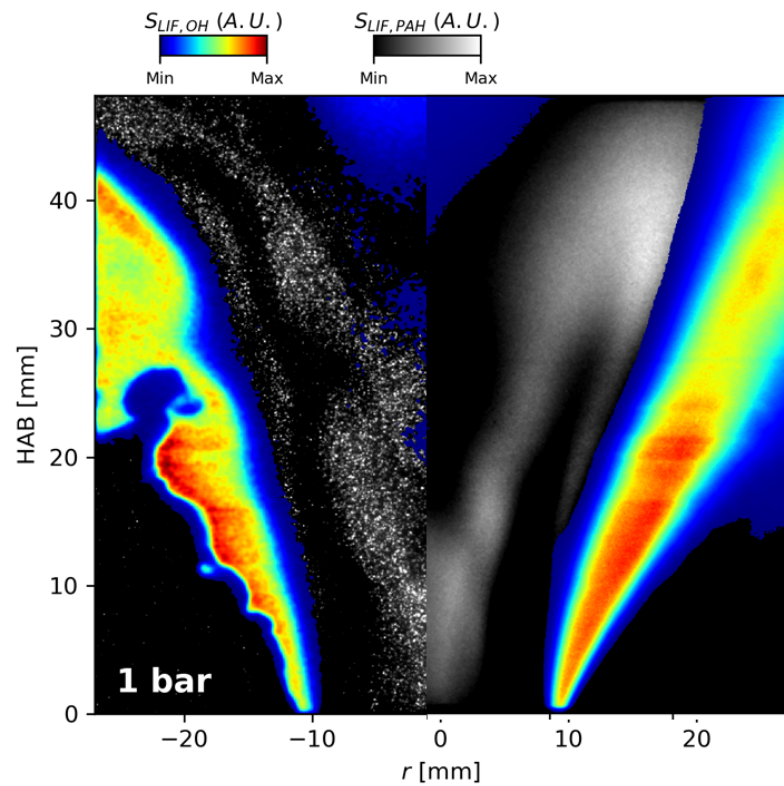


Fig. 6 Combined OH-PLIF (jet colors) and PAH-PLIF (grey colors) signals. Left: Instantaneous image. Right: time-averaged distributions

3.3 Correlation between soot, soot precursors and aerodynamics

As highlighted in Fig. 7, the coupling between the results collected with the OH-PLIF, PAH-PLIF, PIV and LII diagnostics provides useful data required to study the correlations between the velocity and PAH concentration on the soot formation.

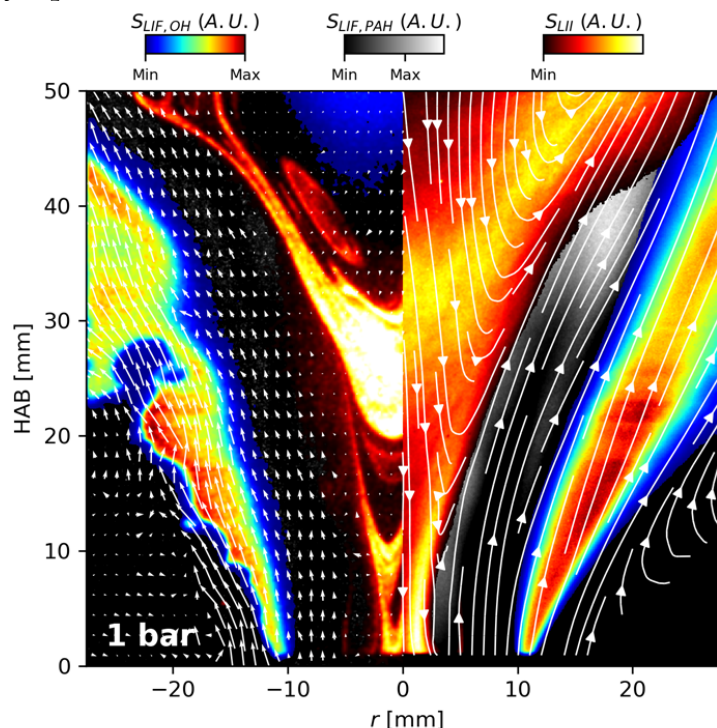


Fig. 7 Superposition of the distributions of OH-LIF (jet color), PAH-LIF (grey color), LII (hot color) with the velocity vectors obtained by PIV. Left: instantaneous images Right: time-averaged distributions

Aromatic precursors generated from the pyrolysis of fuel are transformed in heavy structures which undergo nucleation to finally produce soot particles. Naturally, soot particles when created are either consumed by oxidizing radicals like OH or it suffers a growth by assimilation of other carbonate structures inside IRZ. In our case, the fuel-rich flow injected from the inner tube inhibits the development of the flame towards the centerline where there is a shortage of fresh oxygen due to the circulation of hot burnt gases. As a consequence, it can be observed a lack of intense OH signals in the middle area which could be a sign of an occurrence of combustion. While the internal fuel-rich jet undergoes pyrolysis, the production of aromatic soot precursors is thus detected around the fuel-rich jet. Consumption of PAH occurs rapidly in the vicinity of the flame due to the high concentration of OH at elevated temperature while they subsist in the area between the fuel-rich jet and the boundary of IRZ. In this zone, the soot precursors contribute to a fast production of thin ligaments of nascent soot represented by the LII signals observed on the center of the flow. Indeed, the amount of PAH present can actively participate to the production of nascent soot in a zone where temperature is elevated and the residence time of molecules is high. Nascent soot is then produced at the interface of IRZ in which PAH is consumed. Then, these soot particles are going to be aggregated when they are going to circulate in the center of IRZ. An observation of

Fig. 7 denotes the presence of intense LII signals in this zone which highlights the production of large pockets of soot and emphasizing the transport of mature particles from the higher HAB region towards the centerline of the flame. Weak concentration of OH is also spotted in the central region as this radical subsists into the high-temperature burnt gases transported in IRZ. Of course, the correlation between velocity, PAH concentration and soot production are not sufficient to quantify soot formation, which may also be subject to other physical parameters. Previous studies have shown that the formation of soot particles is promoted not only by chemical mechanisms ensuring a conversion of PAH molecules into soot particles, but also by specific local velocity and strain rate conditions. For instance, coupled LII and PIV measurements have previously been performed on turbulent non-premixed co-flowing ethylene/N₂ jet flames (Narayanaswamy & Clemens, 2013). It was observed that optimal conditions of soot inception are produced for velocities of about 3 m/s and strain rates of 700 s⁻¹, regardless of the jet exit velocity. To better understand the link between soot formation and the parameters mentioned above, spatial correlations between soot aggregates, soot precursors, velocity magnitude and strain rate are displayed on an instantaneous image on Fig. 8.

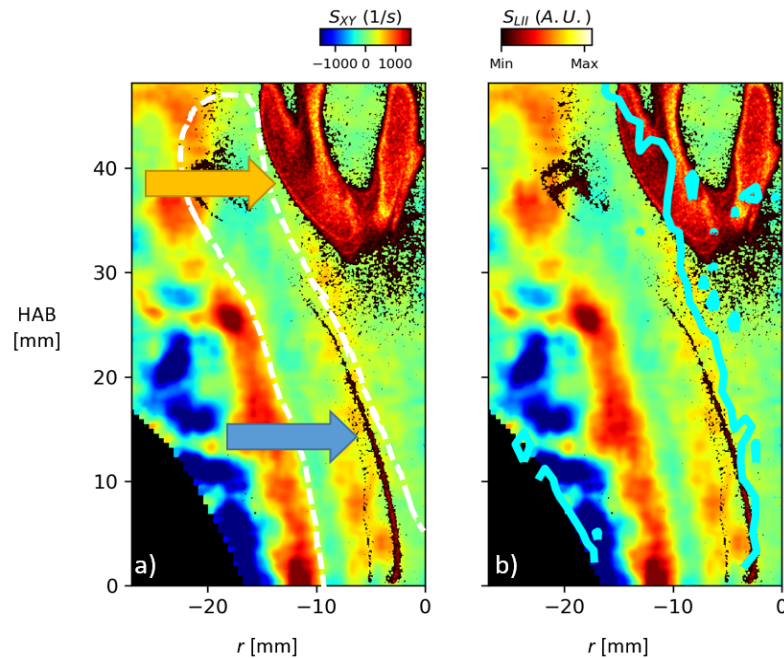


Fig. 8 Example of correlation between strain rate (background) and soot aggregates (hot colors) with soot precursor localization (left, dotted line) and 0 m/s iso-values (right, cyan line)

The local rate of strain has been beforehand calculated thanks to the velocity field measured by the PIV diagnostic, and is defined as following:

$$S_{xy} = \frac{1}{2} \left(\frac{\partial \vec{U}}{\partial \vec{y}} + \frac{\partial \vec{V}}{\partial \vec{x}} \right), \quad (3)$$

- U and V represent the velocity vector components, oriented towards $\vec{x}$ (horizontal) and $\vec{y}$ (vertical) respectively.

Due to the high number of parameters considered, Fig. 8 is split into two diagrams. The left one (Fig.8a) shows the S_{xy} 2D distribution associated with soot aggregates (hot colors) and soot precursors distributions, whose borders are plotted (dotted white line). In the right one, strain rates are displayed with soot aggregate distributions, streamlines (displayed in white) and a thick cyan line denoting the 0 m/s iso-value.

The 2D distribution of local strain rates highlights the formation of three specific shear layers. The external shear layer is located between the co-flow and the outer fuel-rich jet while a shear layer between the outer and the inner jets takes place. These shear layers are characterized by S_{xy} values of about 1000 s^{-1} in the lower part of the flame. The last shear layer, resulting from the velocity gradients between the fuel-lean jet and IRZ, is called here internal shear layer. Less intense strain rate values, from 0 to 500 s^{-1} approximately, are observed in this shear layer.

Regarding the soot spatial distributions, two specific areas are observed distinguished. First, soot aggregates are detected downstream the flame, in the IRZ (yellow arrow), and distributed as a thick pocket. An analysis of successive single-shot images of LII shows that these soot particles occur in this area very intermittently. Moreover, other aggregates are also localized in the lower part of the flame under the form of thin filaments (pointed on Fig. 8a by a blue arrow). These filaments are located at the interface between the inner fuel-lean jet and IRZ. This typical morphology barely differs from one instantaneous frame to another. Given information provided by their localization and PIV measurements, it can be argued that the thick pockets are composed of mature aggregates recirculating towards the center of the flame, while soot aggregates localized in ligaments are nascent. Consequently, only these soot filaments are to be considered in order to try to understand the initial mechanisms governing their production.

We can observe that the filament of soot systematically takes place within the PAH fluorescence region. Besides, soot filaments are also located near as the 0 m/s iso-value, and follows the internal shear layer, where strain rates reach values about 500 s^{-1} , values closed to those found by Narayanaswamy and Clemens. In other words, the region of nascent soot emerges as being a region where all conditions of high residence time, high temperature, high strain rate and existence of soot precursors are favorable to the formation of soot. More interestingly, no soot particles appear elsewhere into the flame, when one of the conditions listed above is not fulfilled. For instance, along the external layer of the PAH region, where the gas axial velocity is much larger

(from 5 up to 15 m/s) and strain rates are more intense (about 1000 s^{-1}). In the same way, no nascent soot is visible inside the PAH region located just downstream the soot filament area (near $r = -10\text{ mm}$ & $HAB = 35\text{ mm}$). Even though axial velocities are low, the strain rate in this zone is nearly reduced to 0 s^{-1} .

4. Conclusion

In the current work, a phenomenal characterization of the mechanisms governing soot formation and oxidation was performed on a stratified swirled premixed ethylene/air flame at atmospheric pressure. To this end, a combination of various optical diagnostics was used for imaging the soot volume fraction (LII), the soot precursors (PAH-PLIF), the flame structure (OH-PLIF) and the aerodynamics (PIV). This combined information allowed the study of the global flame behaviour, and highlighted correlations between soot aggregates and other scalar parameters, helping for the understanding of soot formation mechanisms in this type of combustion. The flame under study presents the shape of a typical swirled stratified flame, featuring a stabilizing recirculation zone. An internal PAH fluorescence signal is visible around the inner fuel-rich jet, suggesting that soot precursors result from the thermal decomposition of ethylene fuel. No PAH fluorescence is observed along the outer fuel-lean flow. Soot aggregates are mainly distributed in two specific regions: A first one, located within the PAH area, along the shear layer between the inner fuel-lean jet and IRZ, produced inception soot arranged in thin filaments. The second area corresponds to IRZ, where soot particles are distributed in thick pockets. Velocity measurements recorded by the PIV diagnostic confirms this observation. In other words, soot filaments are composed of nascent soot in zone in which velocity is low giving then a high residence time for molecules, while aggregates constituting of thick pockets are mature soot emanating from the presence of large moving gas structure which enhance the transport at low velocities of particles soot already produced inside IRZ. In addition to visual observations, PIV measurements were also exploited to calculate the local strain rate, a parameter considered to play a major role in the formation of soot particles. Correlations were established between nascent soot filaments, soot precursors, local velocities and local strain rates. Observations showed that the localization of incipient soot occurs under the combined action of local existence of soot precursors, long residence time, and high strain rate values, which agrees with the assumptions commonly made in soot models. On the contrary, the absence of one of these conditions prevents the production of soot in the flame.

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