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Hugo Pers, Andrea Aniello, Frédéric Morisseau, Thierry Schuller. Autoignition: a flashback initiation mechanism in hydrogen-enriched laminar premixed burners. 11th European Combustion Meeting, CORIA, Apr 2023, Rouen, France. hal-04658074

HAL Id: hal-04658074

<https://hal.science/hal-04658074v1>

Submitted on 23 Jul 2024

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Autoignition: a flashback initiation mechanism in hydrogen-enriched laminar premixed burners

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Abstract

Retrofitting of natural gas-powered premix burners to accommodate hydrogen is limited by the flashback propensity of hydrogen-enriched flames. In this study, flashback initiation is investigated for hydrogen-enriched premixed laminar methane flames. A domestic gas boiler burner is adapted to enable high-speed intensified imaging of its internal part during transition to flashback. A zoom on the location of flashback initiation gives clear evidence for the existence of two distinct regimes. In the first one, the initiation of flashback is due to the upstream propagation of a flame, initially stabilized on the outer surface of the burner wall, through a burner hole. In the second one, spontaneous autoignition of the reactants occurs inside the burner due to their preheating at the contact of the hot internal burner wall, initiating the flashback. Experiments conducted over a large range of hydrogen-enrichment and equivalence ratio confirm these observations. An autoignition Damkohler number Da_i is defined, providing theoretical support to the distinction between the two regimes. These conclusions are finally confirmed in a canonical configuration. They may have significant consequences on the conception of hydrogen-enriched premixed laminar burners.

Introduction

In a global effort to stall climate change, substitution of fossil fuel by carbon neutral alternatives in the heat production sector has become a priority for burner manufacturers. Hydrogen is considered as a promising lead because of its CO_2 -free combustion and its storage capacity [1]. However, its particularly high reactivity hinders the conversion of regular burners to hydrogen [2]. The high laminar burning velocity S_L , the small quenching distance, as well as the tendency to autoignite of hydrogen-air mixtures were shown to challenge the switch from natural gas to H_2 powered systems in industry [1].

One of the main issues in premixed systems, is the propensity of hydrogen flames to flashback inside the burner which raises important safety issues. The inaugural "critical velocity gradient" theory from Lewis [3] aimed at identifying flashback limits of laminar premixed flames. However, this theory does not account for preferential diffusion, flame stretch and coupled heat transfer with the walls. This latter aspect is particularly important for small flames stabilized on perforations [4] like domestic boilers. In particular, the temperature of the burner wall can exceed 1000 K, eventually leading to a large preheating of the reactants in contact with the metallic walls that can trigger flashback for high hydrogen hybridization rate. The typical hole size of these burners being close to the quenching distance of hydrogen flames [5], its reduction generally prevents the propagation of the flame through the hole immediately after ignition when the burner is cold.

Yet, recent observations [2, 6] revealed that in many

cases flashback takes place after a thermal transient state, with the burner wall temperature slowly increasing up to a particular threshold value defined as $T_{w,f}$. Depending on the equivalence ratio and the hydrogen power hybridization rate PH_2 , i.e. the fraction of power originating from the combustion of hydrogen, this threshold could exceed 1100 K. In these cases, the critical velocity gradient theory was shown to be inadequate to predict the flashback limits.

These high temperature flashback thresholds, associated with the very short autoignition delay of hydrogen above the crossover temperature (around 950 K, see [7]), encourage the examination of a potential autoignition of the combustible mixture inside the burner, in the vicinity of the hot walls. The objective of this study is to explore the possibility of autoignition as a flashback initiation mechanism for hydrogen-enriched mixtures with a set of experiments.

The paper is organized as follows. The experimental setup is first presented. Flashback initiation and its dynamics are then examined inside the burner, for industrially relevant cases with wall temperatures below and above the hydrogen crossover temperature. Two regimes of flashback are unveiled and analyzed. The observations made in the industrial burner are then confirmed on a different canonical setup with a simplified burner geometry. Finally, conclusions clarify the origin of flashback in H_2 -enriched multi-perforated laminar premixed burners.

Materials and methods

Figure 1 shows a schematic of the experimental setup. A cylindrical burner is fixed on a converging nozzle above a 100 mm diameter cylindrical plenum.

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Proceedings of the European Combustion Meeting 2023

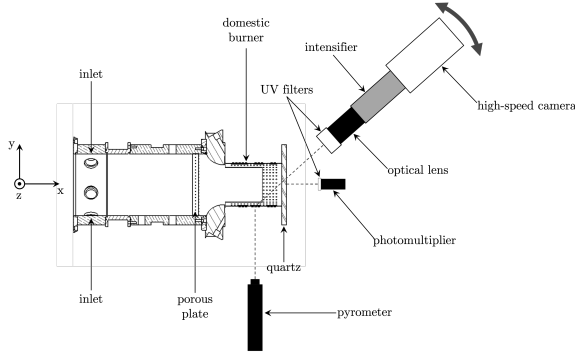


Figure 1: Top view of the experimental setup. The z -axis corresponds to the normal to the (x, y) plane.

The plenum is supplied with methane-hydrogen/air mixtures at atmospheric pressure and temperature. The setup is positioned horizontally to ease the optical access inside the burner. The metallic top of the burner has been replaced by a transparent 8 mm thick disk made of fused silicate. Details about the experimental setup can be found in [6].

A side view of the burner with images during the transition to flashback are presented in Fig. 2. After ignition, the burner wall temperature increases until reaching a threshold at which flashback occurs. This burner has the same characteristics as standard technologies used in domestic boilers. Its height and diameter are respectively 91 mm and 70 mm. It is formed by a 0.6 mm sheet made of refractory steel, bent and welded to form a cylinder. It is perforated with a regular pattern made of holes and slits. The diameter of the holes is 0.7 mm and the slits are 0.1 mm large and 3.5 mm long. A detailed burner description is available in [6].

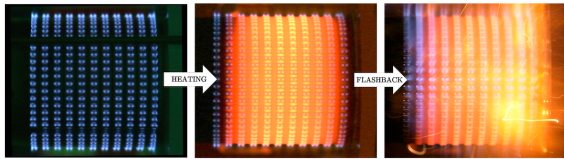


Figure 2: Sequence leading to flashback on the cylindrical burner recorded by a color-camera at 60 frames per second. Conditions are $\phi = 0.78$, $PH_2 = 65\%$, $P = 3$ kW.

The bulk velocity U_b in each hole is deduced from the total mass flow rate through the burner:

$$U_b = \frac{\dot{m}_h}{\rho_u A_h} = \frac{\sigma \dot{m}_p}{\rho_u A_h N_p} \quad (1)$$

where \dot{m}_h , \dot{m}_p , ρ_u , N_p , σ and A_h are respectively the mass flow rate through a single hole, the mass flow rate through a patch, i.e. here two holes and two slits [2], the density of the preheated gases at T_u , the total number of patches, the ratio of a hole surface area over a patch surface area, and the surface area at the outlet of a single hole.

Flashback initiation is scrutinized using a Phantom V1612 camera coupled with a Lambert HiCATT intensifier and a Micro-Nikkor 105 mm UV lens, equipped with a 305-315 nm band pass filter.

The camera can be aligned with the burner axis or set at a 35° angle with respect to the burner axis to focus on the initiation region, as depicted in Fig. 1. The camera and the setup are kept aligned within the (x, y) plane. Additionally, a Hamamatsu H10722-04 photomultiplier (PM) equipped with a 305-315 nm band pass filter is used to record the total light emission during flashback.

The external burner wall temperature T_w is obtained with a bichromatic infrared pyrometer (FLUKE Endurance Series). The hot burner walls cause the temperature of the combustible mixture leaving the burner holes T_u to be higher than the inlet gas temperature $T_{in} = 300$ K. The preheat temperature T_u is determined using the method described in [2, 6, 8].

The analysis concentrates on flashback scenarios in which the temperature T_w of the metallic burner envelope gets sufficiently high, surpassing 500 K. Under these conditions, flames stabilize above the cold surface of the burner and the temperature T_w of the burner rises until a particular threshold temperature $T_{w,f}$ is reached, at which flashback takes place [2]. Events of flashback right after the ignition of a cold burner are not considered in this work. The experimental procedure is detailed in [6].

The preheat temperature T_u of the reactants leaving the burner holes is used to determine both the bulk velocity of the reactants flowing through a single hole U_b and the laminar burning velocity S_{L,T_u} . For the studied burner, the ratio of preheat temperature over wall temperature T_u/T_w turns out to be quasi-constant from an operating point to another, leading to the following empirical expression valid at $P \approx 3$ kW and for T_w between 950 K and 1100 K:

$$T_u \simeq 0.74T_w \quad (2)$$

This relation is applied in the following sections to estimate T_u from measurements of T_w . The origin of flashback of hydrogen-enriched flames is now investigated using high-speed imaging.

Propagation vs autoignition flashback regimes

Observation of the initiation location of flashback with the high-speed camera aligned with the axis of the burner reveals that initiations occurs mostly close to the weld [6]. It was shown that at this location the reactants reach higher preheat temperatures due to a longer residence time, which results in faster flames and a hotter wall surface in this region, hence making it a preferential flashback zone.

The high-speed camera is placed off axis as in Fig. 1, focusing towards the area of preferential flashback initiation, around the weld. Operating conditions leading to two different scenarios are investigated. They are defined as case A and case B. Conditions are respectively $\phi = 0.6$, $PH_2 = 100\%$, $P = 3$ kW for case A and $\phi = 0.75$, $PH_2 = 67\%$, $P = 3.25$ kW for case B. Associated wall and preheat temperatures are detailed in Tab. 1.

These operating conditions are chosen because of the almost constant ratio of bulk velocity over laminar burning velocity $(U_b/S_L)_{T_u} \simeq 0.6$ calculated at preheat temperature T_u , which avoids considering a potential hydrodynamic imbalance between the two cases, and most importantly because of the important gap between wall temperatures at which flashback takes place, respectively $T_{wd,A} = 953$ K and $T_{wd,B} = 1053$ K.

Table 1: Comparison, for cases A and B, of equivalence ratio ϕ , H_2 -hybridization power rate PH_2 , global power, wall temperature at flashback $T_{w,f}$, T_u obtained from Eq. (2), and ratio of bulk velocity over laminar flame speed $(U_b/S_L)_{T_u}$.

	ϕ	PH_2	Power (kW)	$T_{w,f}$ (K)	T_u (K)	$(U_b/S_L)_{T_u}$
Case A	0.60	100	3.0	953	705	0.64
Case B	0.75	67	3.25	1058	783	0.62

These temperatures are also close to the crossover temperature $T_c \simeq 950$ K for typical H_2 /air mixtures [7], which is defined as the temperature at which chain-branching reaction rate equals the recombination reaction rate [7, 9]. Approaching this value initiates a dramatic decrease of the autoignition delay time, typically dropping from 100 ms to 0.5 ms between 900 K and 1000 K [6]. A short ignition delay time could potentially lead to autoignition, given a sufficiently long time of residence of reactants beneath the wall.

Figure 3 displays flashback initiation sequences for case A and case B, side-by-side, recorded with a sampling rate of 16 kHz. Initiation locations are emphasized on the second row with close-up inserts that clearly show two different locations of flashback: inside a hole in case A (propagation regime), and directly from the inner wall along the weld in case B (autoignition regime). Experiments in each case were repeated 10 times to exclude the possibility of a marginal phenomenon, each time leading to the same conclusion, the only difference being the location of the hole where flashback is initiated or the exact location of autoignition below the wall.

These observations indicate that when the burner temperature reaches temperatures higher than $T_c \sim 950$ K, two flashback mechanisms can take place. The first one is labelled FB-H for FlashBack originating from Hydrodynamic imbalance, and characterizes a flame stabilized above the perforated burner that can propagate upstream through a hole. This is the well known flashback mechanism that takes place when

the wall temperature remains below the $T_c \sim 950$ K threshold. The second one, defined as FB-AI for FlashBack originating from AutoIgnition, is only observed for higher wall temperatures, in which case the flammable mixture spontaneously ignites next to the hot inner burner wall.

To confirm these findings, a set of operating conditions that allow for an accurate, reproducible identification of the flashback initiation point is scrutinized. Points were selected to keep the ratio between the bulk velocity and the laminar burning velocity at the preheated temperature, $(U_b/S_L)_{T_u}$, almost constant, around 0.6. As the wall temperature $T_{w,f}$ at flashback and thus T_u strongly depend on the chosen operating point, it is difficult to predict the ratio $(U_b/S_L)_{T_u}$ beforehand. Therefore, conditions that yield $(U_b/S_L)_{T_u} \simeq 0.6$ were identified afterwards. The list of the examined operating conditions, wall temperatures T_w and preheated gas temperatures T_u , as well as the type of flashback observed are summarized in Tab. 2. These conditions were also chosen so as to be relevant to the industrial operating range.

Table 2: Characteristics and initiation regimes of investigated conditions.

PH_2 (%)	ϕ	Power (kW)	$T_{w,f}$ (K)	$T_{u,f}$ (K)	$(U_b/S_L)_{T_u}$	$Da_i = \frac{u}{\tau_i}$	Initiation location
100	0.6	3	953	705	0.64	0.13	Hole
100	0.61	3	940	696	0.63	0.03	Hole
88	0.67	3	953	705	0.61	0.14	Hole
85	0.70	3	950	703	0.59	0.11	Hole
85	0.72	3	937	694	0.57	0.03	Hole
72	0.71	3	1050	777	0.59	2.8	Wall
68	0.73	3	1055	781	0.59	3.0	Wall
67	0.75	3	1058	783	0.57	2.6	Wall
67	0.75	3.25	1053	779	0.62	2.3	Wall
67	0.75	3.5	1058	783	0.67	2.3	Wall
65	0.78	3	1067	790	0.55	2.8	Wall
65	0.78	3.5	1079	799	0.63	2.7	Wall
65	0.78	5	1081	800	0.89	1.9	Wall

These experiments enable to discriminate two categories: a first group of hole-initiated flashbacks occurring at wall temperatures $T_w < 955$ K and a second group of flashbacks initiated directly from the burner wall when $T_w > 1050$ K. These results highlight the role of the crossover temperature T_c , above which a sudden drop of the autoignition delay is observed for hydrogen-enriched premixed flames, eventually triggering flashback for flames stabilized above a burner when the walls become too hot. It is worth emphasizing that this would not be the case for operation with natural gas or with other hydrocarbon fuels as the crossover temperature is specific to hydrogen [7], although the temperature dependency of hydrocarbon ignition delay do change above 1400 K [10]. A criterion is now considered to investigate conditions leading to autoignition.

Transition criterion for autoignition

To determine whether autoignition can take place, the autoignition delay time τ_i needs to be evaluated

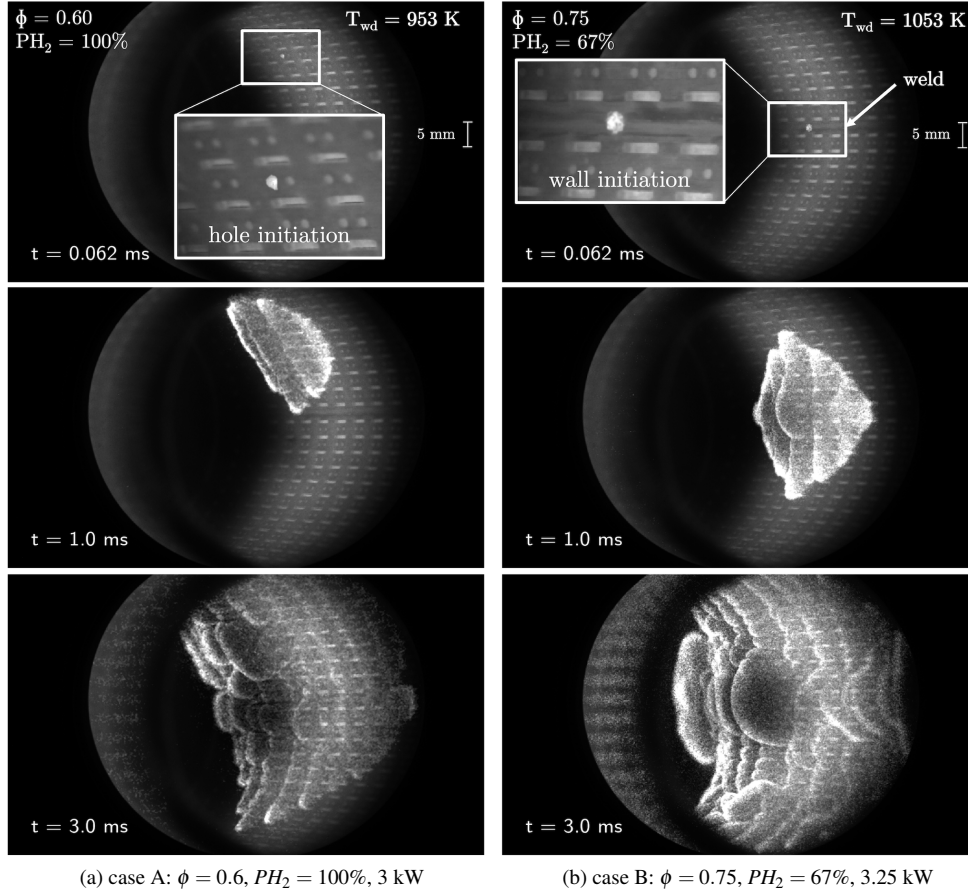


Figure 3: High-speed imaging of flashback initiation and propagation, for two regimes of flashback: hole initiation (a) and wall initiation (b). T_{wd} is the temperature at the weld location.

against the residence time of the reactants close to the wall. The residence time is calculated as $\tau_r = L_w/U_b$, where L_w is the width of the weld and U_b is the bulk velocity of the reactants in burner holes calculated at the preheat temperature T_u . The ratio of τ_r/τ_i corresponds to a Damkohler number, as it compares a flow time scale and a chemical time scale:

$$Da_i = \frac{\tau_r}{\tau_i} = \frac{L_w}{U_b} \frac{1}{\tau_i} \quad (3)$$

Figure 4 plots the autoignition Damkohler number Da_i as a function of the velocity ratio $(U_b/S_L)_{T_u}$, for the conditions presented in Tab. 2. A large gap is observed between flashback initiated through a hole, regime FB-H, and flashback initiated at the burner wall, regime FB-AI. The residence time of the reactants next to the hot wall being larger than the autoignition delay time strongly supports the scenario of an autoignition-driven regime FB-AI. Conversely, small values of Da_i for FB-H conditions tend to discard this mechanism for flashback initiation.

Coexistence of flashback regimes

Figure 5(a) maps the different flashback regimes observed for the cylindrical burner as a function of

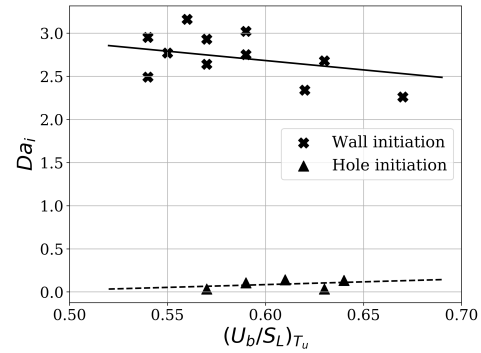


Figure 4: Autoignition Damkohler number $Da_i = \tau_r/\tau_i$ as a function of $(U_b/S_L)_{T_u}$ in regimes FB-H (triangle markers) and FB-AI (square markers).

equivalence ratio ϕ and hydrogen hybridization rate PH_2 , for $P = 3$ kW. Stable operation (bottom left corner) can only be obtained for lean conditions with a limited H_2 hybridization rate.

In the upper right corner in Fig. 5(a), flashbacks occurring instantaneously after ignition (INSTANT FB) can be observed, for highly enriched mixtures close to stoichiometry. The domain in between the STABLE and

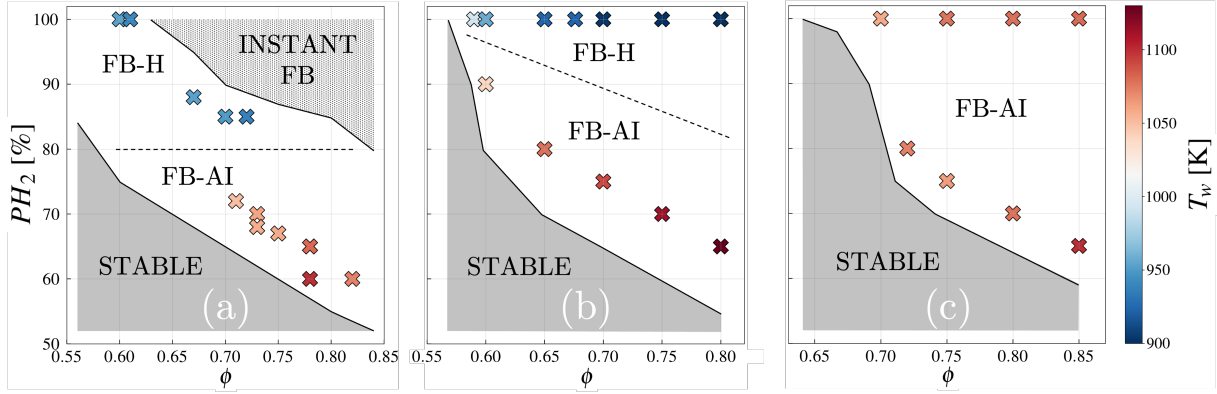


Figure 5: Stabilization maps as a function of equivalence ratio and hydrogen hybridization power rate. Studied flashback conditions are colored by the wall temperature threshold $T_{w,f}$. The power density through perforations remains almost constant at $P_s \approx 4 \text{ MW/m}^2$. (a) Cylindrical hole-perforated burner, $D = 0.7 \text{ mm}$, $P = 3 \text{ kW}$. (b) Slit-perforated plate B, $D_B = 0.37 \text{ mm}$, $P = 0.5 \text{ kW}$. (c) Slit-perforated plate C, $D_C = 0.32 \text{ mm}$, $P = 0.5 \text{ kW}$.

INSTANT FB regions corresponds to flashback taking place at a given temperature threshold $T_{w,f}$, during the thermal transient after ignition. In this case, the two identified regimes appear separated. The hydrodynamic FB-H regime essentially occurs for highly H_2 -enriched lean mixtures, when the autoignition-driven FB-AI regime is found for richer conditions featuring a smaller hydrogen content. The delimitation between these flashback regimes is mostly related to the burner wall temperature at flashback $T_{w,f}$.

It is likely that a hole size reduction, while keeping a constant power density, would quench even the most reactive flames and prevent upstream propagation through the holes. In such cases, autoignition could potentially become the only regime of flashback.

To verify this conjecture, additional experiments are conducted on a different setup with slit-perforated plates. The objective is to prove that the autoignition regime that has been identified in the cylindrical industrial burner is independent of the burner geometry, i.e. independent of a specific hole and slit pattern and independent of a welded part. The injection setup in Fig. 1 is now placed vertically and the cylindrical burner is replaced by a slotted plate, an example of which is shown in Fig. 6.

The choice of slits instead of holes is made to get closer to 2D configurations that are currently being simulated in a companion study. Plates are squares of 59 mm side, 6 mm thick, made of the same refractory steel as the cylindrical burner. The perforation pattern is laser cut. Two plates are examined here. Plate B features a slit width $D_B = 0.37 \text{ mm}$, and plate C has a slit width $D_C = 0.32 \text{ mm}$. In both cases, slit spacing is $W = 2 \text{ mm}$, row spacing is $H = 4 \text{ mm}$ and slit length is $L = 6 \text{ mm}$. The hydraulic diameter of plate B slits is $D_B^h = 0.7 \text{ mm}$, which is exactly the diameter of the cylindrical burner holes. Plate C slits hydraulic diameter is $D_C^h = 0.6 \text{ mm}$. Transition to flashback is investigated at $P = 0.5 \text{ kW}$ for both plates, corresponding to the same power density

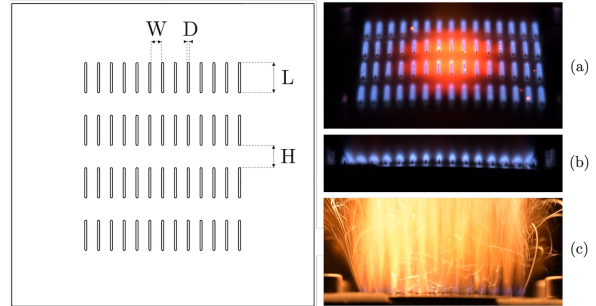


Figure 6: On the left: Example of slit-perforated plate. On the right: (a) Upper view of the plate B in conditions $\phi = 0.68$, $PH_2 = 97\%$, $P = 1.5 \text{ kW}$. (b) Side view of plate in the same conditions. (c) Example of flashback on a plate with $D = 0.5 \text{ mm}$ (not presented here) in conditions $\phi = 0.68$, $PH_2 = 97\%$, $P = 2 \text{ kW}$.

$P_s \simeq 4 \text{ MW/m}^2$ as the cylindrical burner operating at $P = 3 \text{ kW}$.

Stabilization maps for plates B and C are presented in Figs. 5-(b) and 5-(c) respectively. The wall temperature is measured at the hottest point, between two slits in the center of the plates. As for the cylindrical burner in Fig. 5-(a), stable combustion is only achieved at the bottom left corner in Figs. 5-(b,c), but instantaneous flashback (INSTANT FB) after ignition disappears for plates B and C, meaning that a non-preheated flame is never able to move upstream through a cold plate for these slit sizes. Nevertheless, combustion remains unstable in the upper right corner in Figs. 5-(b,c) with flashback taking place during the thermal transient state. Regimes leading to flashback are discriminated using the Damkohler number Da_i defined in Eq. (3), and by choosing $Da_i = 1.5$ as the threshold above which FB-AI regime is beyond doubt. Equation (2) is used to estimate the preheat temperature T_u and deduce the bulk velocity U_{b,T_u} in the slits. The residence time is defined as here $\tau_r = W/U_{b,T_u}$, where H is the space between two slits (Fig. 6).

For plate B in Fig. 5-(b), the FB-H domain still exists but is substantially reduced, as the flashback limit of the stable domain is always due to FB-AI except for pure hydrogen flames $PH_2 = 100\%$. Plate C presents a yet different flashback domain. All flashbacks occur at wall temperature $T_w > 1050$ K, and FB-AI appears to be the only triggering mechanism for all equivalence ratios and all hybridization rates. Indeed, FB-AI initiation occurring even in the most reactive case ($100\%H_2$ at the highest considered equivalence ratio), which is the most likely to be able to flashback hydrodynamically, no operation condition appears to follow a FB-H flashback mechanism.

These experiments confirm that reducing the size of the burner perforations eventually leads to the disappearance of the standard hydrodynamic flashback regime FB-H, in which case autoignition becomes the only flashback mechanism. This endorses the fact that slit size reduction alone is insufficient to prevent flashback, and leads to a rethinking of how premix burners should be adapted to hydrogen hybridization.

Conclusion

Flashback in H_2 -enriched laminar premixed burners has been investigated, exploring the possibility of autoignition events as a function of equivalence ratio, hydrogen hybridization rate and power with a high-speed intensified imaging system inside a burner equipped with a large optical access.

Experiments focusing on the flashback initiation region produce explicit evidence for the existence of two regimes determined by the burner wall temperature. The FB-H regime, FlashBack driven by Hydrodynamics, was observed for mildly hot burner walls, when the wall temperature remains below 950 K. In this case, the initiation of flashback occurs inside a burner hole because of the imbalance between the flow velocity and flame propagation leading to a flame propagating upstream through the hole. In the second FB-AI regime, FlashBack driven by AutoIgnition, spontaneous autoignition originates away from any hole at the inner burner wall when its surface is hot enough, around 1050 K. An autoignition Damkohler number Da_i has been introduced, comparing a residence time of reactants flowing along the hot wall and the autoignition delay time. Comparison of Da_i for operation at constant $(U_b/S_L)_{T_u}$ enables to discriminate between FB-H and FB-AI regimes, corroborating the existence of the two distinct mechanisms.

Additional experiments on canonical slits-perforated plates confirmed that the phenomena observed are not specific to the burner considered. All the more, it is shown that reducing the dimensions of the holes or slits cannot prevent flashback, but that the FB-AI mechanism takes over the FB-H standard mechanism and eventually becomes the only flashback mechanism.

These conclusions may lead to rethink the development of hydrogen-enriched premixed laminar burners. Given the significantly high wall temperatures

produced by hydrogen-enriched premixed flames, the risk of autoignition should be considered making hole size reduction ineffective to hinder flashback.

Acknowledgments

This project received funding from SERMETA, from the ANRT (grant n° 2021/1069), from ERC SCIROCCO (grant 832248) and from the Horizon 2020 COEC program (grant 952181). The development of the experimental setup would not have been possible without the help of S. Cazin, G. Albert, S. Lun and L. Mouneix.

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