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Full length article

Stabilization and emissions characteristics of $\rm CH_4-H_2$ blends in a premixed jet stabilized combustor

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HIGHLIGHTS

- + $\rm CH_4/\rm H_2$ admixtures from 0–100 % $\rm H_2$ and lean equivalence ratios.
- Two main recirculation zones, where the PRZ has a higher recirculation ratio than the CRZ.
- Temperature, CO and NO mole fraction measurements in the reaction zone.
- Higher NO for higher H_2 % for the same ϕ , but lean stable limit with higher H_2 %.
- Higher NO for higher H_2 % for the same ϕ , but lean stable limit with higher H_2 %.
- Shift in reaction zone stabilization mechanism from autoignition to flame propagation when H_2 % increases or ϕ increases.

ARTICLE INFO

Keywords: Jet recirculation Flameless/MILD Autoignition Emissions H₂ admixture

ABSTRACT

Low emissions and fuel flexibility are two important criteria required for gas turbine combustors to facilitate the energy transition to low-carbon fuels for propulsion and power applications. A jet-stabilized combustor, having both these characteristics, was operated with CH₄-H₂ fuel mixtures with H₂ varying from 0 to 100 % and with varying equivalence ratios (ϕ). Comprehensive measurements were carried out of the velocity field using Particle Image Velocimetry (PIV), temperature and gas composition by traversing probes in the chamber, and flame topology using chemiluminescence imaging. The flow field in this combustor consists of a jet that undergoes recirculation, generating Central and Peripheral Recirculation Zones (CRZ and PRZ). The recirculation ratio in the PRZ is found to be twice that of the CRZ. Increasing H_2 % for the same ϕ leads to higher NO_x. Ultra-low ϕ flames could be stabilized only at H₂ \geq 50 %, which in turn leads to low NO_x due to low adiabatic flame temperatures. The combination of temperature, gas composition (CO/NO), and chemiluminescence images is used to identify the extent and location of the reaction zone. Distributed reaction zones, stabilizing at around 30 % of the length of the chamber, are achieved at lean conditions, whereas an increase in H_2 % makes the reaction zone more compact and shifts upstream towards the burner head. Flame kernels are extracted from the instantaneous chemiluminescence images, and probability distribution functions for their aspect ratio and axial location are constructed. It is seen that reducing ϕ leads to low aspect ratio kernels that tend to occur further downstream, whereas increasing H₂ % leads to higher aspect ratio kernels, stabilizing upstream. These flame kernel statistics are also used to identify ignition modes (autoignition/flame propagation) for varying fuel H2 % and inlet ϕ based on a hypothesis of flame stabilization mechanisms.

1. Introduction

Hydrogen has emerged as a promising energy carrier for reducing carbon emissions from combustion devices, aligning with global efforts to mitigate climate change [1,2]. However, the direct substitution of conventional fuels with hydrogen presents several technical challenges, including an increased risk of flashback, combustion instabilities, and potentially higher NO_x emissions [3,4]. Moreover, the near-term availability of green hydrogen is projected to be limited, potentially insufficient to meet current energy demands [5]. To address these constraints while still leveraging the benefits of hydrogen, a viable strategy involves blending conventional fuels with hydrogen

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Nomenclature	L_c Characteristic length scale
$\begin{array}{l} \chi \ \ \mbox{Mole fraction} \\ \Delta t \ \ \mbox{Time interval between the successive PIV frames} \\ \delta_f \ \ \ \mbox{Laminar flame thickness} \\ \epsilon \ \ \mbox{emissivity} \\ \phi \ \ \mbox{Equivalence ratio} \\ \sigma \ \ \mbox{Stefan-Boltzmann constant} \\ \sigma_u \ \ \mbox{standard deviation of velocity} \\ \tau \ \ \mbox{Time scale} \\ AR \ \ \mbox{Aspect Ratio} \\ BPTRZ \ \ \mbox{Broadened Preheat Thin Reaction Zone} \\ COM \ \ \mbox{Center of Mass} \\ CRZ \ \ \mbox{Central Recirculation Zone} \\ D \ \ \mbox{Diameter of fuel/air mixture nozzle (mm)} \\ d \ \ \mbox{Diameter of fuel nozzle (mm)} \\ FOV \ \ \mbox{Field of View} \\ h \ \ \mbox{Convective heat transfer coefficient} \end{array}$	LIF Laser Induced Fluorescence N Number of samples PDF Probability Density Function PIV Particle Image Velocimetry PLIF Planar Laser-Induced Fluorescence PRZ Peripheral Recirculation Zone PSR Perfectly Stirred Reactor R_0 Radius of combustion chamber cylinder r_{wall} Radial distance from wall RR Recirculation Ratio S_L Laminar flame speed T.I Turbulence Intensity UV-RAS Ultraviolet Resonant Absorption Spectroscopy WSR Well Stirred Reactor X Axial distance from burner head Y Distance along the diameter of the chamber

to incrementally reduce carbon footprints. While hydrogen admixing does reduce the carbon emissions, there is still an issue of high NO_x emissions. The flameless combustion regime is one possible combustion strategy that can be exploited as it has good potential for the reduction of NO_x emissions, which makes it an attractive technology for gas turbine engines [6]. It is also associated with autoignition as the primary stabilization mechanism, as opposed to flame propagation [7,8], which makes it important to analyze the ignition mechanism of different fuel admixtures under varying operating conditions for optimal design.

A jet-stabilized combustor is a configuration within which Flameless/MILD (Moderate or Intense Low-oxygen Dilution) Combustion regime can be achieved in premixed/partially premixed fuel-air mixtures [9-11]. This configuration consists of reactants entering the combustion chamber through high-velocity jets at speeds that are typically much higher than the turbulent flame speed. These jets are located off the axis of the combustor such that a strong recirculation zone is established, thereby supplying hot combustion products back to the reactant stream, which causes it to ignite and thus stabilize a reaction zone in the chamber. The mixing of reactants and recirculated hot products provides suitable conditions, i.e., high temperature of reactants and dilution of O₂ content by the addition of diluents, leading to Flameless/MILD combustion regime [12,13]. Studies such as [9] and [10] experimentally analyzed CH₄-H₂ flames in a jet stabilized combustor, with a maximum H₂ admixture of 40 % (by volume) with natural gas. The authors demonstrated the suitability of Flameless combustion at gas turbine conditions by preheating the air and operating at 20 bar. A combination of OH-PLIF and OH* chemiluminescence was used to characterize the reaction zone and emissions were measured at the exhaust using an exhaust gas analyzer. Low NO_x emissions were achieved when the reaction zone was homogeneously distributed over a large volume, and the equivalence ratio (ϕ) was < 0.5. Further, it was postulated that by increasing jet velocity, the mixing between fresh fuel/air and recirculated burnt gases was enhanced and the residence time was reduced. [14] investigated the flame stabilization mechanism in a single jet stabilized combustor, operating on a CH₄ air mixture at $\phi = 1$, T_{in} = 473 K, Re = 5640. This resulted in a visible, lifted flame which was measured by OH-LIF, PIV, and OH* chemiluminescence, all at 5 kHz, thus yielding time-resolved information of the phenomenon. Vortices were generated in the shear layer, but only on the side where there was enough distance between the jet and the wall. These vortices induce the mixing of the recirculated hot products and fresh reactant stream, leading to a series of events in which the mixture ignites, there is a flame wrap-up, flame expansion, and finally, a merger into the main flame body. While flame propagation was attributed to cause most of the flame expansion, autoignition could not be ruled out. Autoignition was seen to occur through isolated flame kernels at the bottom of the flame body at high frequency, which had a significant effect on the lift-off height. Although the relative weightage of autoignition and flame propagation was difficult to quantify based on these diagnostics, it was clear that the mixing of hot products of combustion and fresh reactants led to the formation of flame precursors, which may enable both flame propagation and autoignition further downstream. Liu et al. [15] investigated the effect of H_2 addition to CH_4 in a mild combustor, which was a jet-stabilized FLOX burner similar to the one in the current study. They explored H₂ mixtures up to 50 % (by volume). The authors presented measurement results from direct imaging by DSLR and OH* chemiluminescence to image the flame topology. Further, the exhaust flue gas temperature and composition were measured using a type B thermocouple and an exhaust gas analyzer, respectively. The combustor was also modeled using a Chemical Reactor Network. Ultra-low NO_x (< 5 ppm) was reported and the addition of H_2 led to a reduction in reaction zone lift-off height. The authors also found it difficult to stabilize a reaction zone above 50 % H₂.

While the above are examples of premixed/partially premixed systems, the literature also encompasses flameless combustion systems wherein the fuel and air are injected through separate streams and interact in the combustion chamber. The operational characteristics of a small-scale combustor were studied by Veríssimo et al. [16]. The combustor was non-premixed, with a central air nozzle surrounded by fuel nozzles. Emissions were measured for $\phi \leq 1.0$, and NO_x first increased, peaked at $\phi = 0.5$ with a value of 9 ppm, and then decreased. On the other hand. CO first decreases and then increases below this value of ϕ . Further studies were done to assess the effect of inlet air velocity [17], thermal power input [18] and air preheat temperature [19]. The effect of increasing air velocity by changing nozzle diameter, i.e., keeping all mass flows constant, resulted in lowering of NO_x and increasing CO. This is attributed to higher entrainment of fuel and recirculating product gases into the air jet, which helps in the onset of flameless combustion. Increasing thermal power implied increasing velocity of all inlet streams while keeping the equivalence ratio constant. This had a significant effect on residence time, thus higher thermal power cases had lower residence time. This led to higher CO at the exit; however, the NO_x remained constant. This was attributed to the suppression of the thermal NO_x pathway under FC regime and it is stipulated that the N₂O pathway may have a more important role.

Ayoub et al. [20] performed measurements on a reverse flow nonadiabatic combustor where all the combusted product gases are recirculated and exhausted through the burner head where fuel and air are injected. There are cooling tubes in the combustor which extract heat to emulate a process load. They performed OH* chemiluminescence imaging and exhaust gas measurements to characterize the reaction zone optically and the resultant net emissions. The furnace was operated with methane as well as methane–hydrogen mixtures by Ferrarotti et al. [21], and they observed that NO_x increased with increasing H₂ % from 0 to 100 %. Decreasing the fuel injector diameter from 25 to 16 mm led to a reduction of NO_x from 175 ppm at 3 % O₂ to 60 ppm at 3 % O₂ for 100 % H₂. By increasing the length of the fuel lance protruding into the chamber from 0 to 50 mm, the NO_x could be further reduced from 60 to 30 ppm for 100 % H₂. This was attributed to the extra residence time available for the air to mix with the exhaust gases before interacting with the fuel stream. The pure CH₄ case showed negligible levels of NO_x.

The jet-in-hot-coflow (JHC) has emerged as a popular choice to emulate conditions created locally in a flameless/mild combustor in the laboratory while allowing for high-fidelity laser diagnostics. Medwell et al. [22] observed that for cases with hot coflow, high levels of formaldehyde (CH₂O) were also seen in the preignition zone, i.e., much upstream of the actual lift-off height. This suggested that MILD combustion conditions lead to a different flame stabilization mechanism. Gordon et al. [23] identified a series of temporally successive phenomena based on previous experiments and numerical simulations that indicated the existence of autoignition. The events are:

- 1. Build up of precursor pool: where the mole fraction of CH_2O increases significantly and that of OH remains low.
- Initiation of reaction: CH₂O and OH mole fractions reach a maximum in a very short period of time.
- Formation of a steady flame: CH₂O peak decreases while the OH peak mole fraction does not change.

The authors took simultaneous OH, CH_2O PLIF, and Rayleigh temperature measurements in a flame of methane in vitiated coflow and identified the existence of these events. They concluded that they observed a high percentage of autoignition events at the base of the lifted flame, and thus, it was highly likely to be the stabilization mechanism for such a flame.

Oldenhof et al. [24] tracked ignition kernels in the Delft jet-in-hotcoflow (DJHC) and analyzed the lift-off behavior. The trends for the sensitivity of lift-off height to jet Reynolds number were completely different for hot and diluted coflow compared to conventional lifted flames. The addition of higher alkanes and increasing the coflow temperature had similar effects, leading to lower axial location where ignitions first occur, higher ignition frequencies, and reduced lift-off height. A quantitative analysis of entrainment of coflow gases into the central jet [25] revealed that the rate at which oxidizer is transported to the shear layer between the jet and the coflow, where mixing occurs, is critical to flame stabilization. Specifically, in the case of the DJHC, there is a positive radial temperature gradient, and an increase in jet velocity leads to increased entrainment of hot coflow, which leads to the initial drop in lift-off height with jet velocity.

To study autoignition, one must be able to identify and distinguish it from flame propagation, which is the stabilization mechanism for a conventional flame. Several researchers have worked on developing criteria for the same. Gordon et al. [7] have developed two indicators based on numerical modelling. The first one is a budget of convection, diffusion, and the reaction of key species wherein it was shown that for a conventional flame, diffusion plays an important role, whereas for autoignition, the convection and reaction terms mostly balance out each other. The second is the build-up of HO₂ prior to autoignition followed by the creation of H, O, and OH, whereas, in conventional premixed flame propagation, all radicals would build up simultaneously. On the other hand, Schulz et al. [26] derive an autoignition index solely based on the chemistry. HO₂ is denoted as a critical intermediate specie in the heat release reactions, and it is found that the consumption occurs through two main reactions: $HO_2 + H \rightleftharpoons OH + OH$ (R6) and $HO_2 + OH \rightleftharpoons H_2O + O_2$ (R8). The relative extent of each reaction turns out to be a good indicator of whether the reaction is stabilized by flame propagation(R6>R8) or autoignition(R8>R6). This criterion is used to identify autoignition events in a turbulent lifted methane-air flame, and it is shown that premixing and opposed mixing mode autoignition are the prominent mechanisms of stabilization in the outer shear layer and at the flame base. Aditya et al. [27] extended the Chemical Explosive Mode Analysis (CEMA) to distinguish between autoignition and flame propagation. The ratio of the non-chemical to the chemical source term ($\alpha = \phi_s / \phi_{\omega}$), projected to the left eigenvector of the Chemical Explosive Mode, indicates the relative importance of diffusion to chemical kinetics. The value of this parameter is used to denote assisted ignition, i.e., flame propagation, where diffusion plays a significant role, autoignition, and local flame extinction. This indicator is used to identify in the Direct Numerical Simulation (DNS) of a reheat combustor that most of the fuel is consumed via the autoignition process.

While jet-stabilized combustors are suitable for low NO_x combustion, there have been limited studies on the nature of the flame stabilization mechanism (autoignition/flame propagation) under varying hydrogen admixture conditions. Further, to the best knowledge of the authors, there are no studies of this configuration providing temperature and gas composition within the reaction zone, leaving much of the detailed reaction kinetics to informed extrapolation or entirely derived from simulations.

This study bridges a critical research gap on premixed jet-stabilized combustors operating on CH₄/H₂ fuel admixtures by providing a comprehensive experimental dataset that includes velocity fields, temperature, composition, and flame chemiluminescence for varying equivalence ratios and H₂ content, varying from 0 to 100 % H₂ by volume, in the fuel mixture. Our extended PIV measurements cover both Central and Peripheral Recirculation Zones, offering a more complete flow field view, which is used to quantify the recirculation ratio based on the detected interfaces. Detailed temperature and composition measurements in the reaction zone provide unprecedented insight into chemical reaction progress within the chamber. We present an innovative approach to analyzing low-fidelity chemiluminescence data to distinguish between autoignition and flame propagation modes and the mode of ignition and stabilization of the reaction zone is observed to shift based on reactant composition. Integrating these multi-parameter analyses offers a unique characterization of this combustor configuration.

2. Experiment setup

2.1. Setup and operating conditions

The experiments were conducted in a jet-stabilized combustor, shown in Fig. 1 the burner head of which consists of 12 nozzles of diameter, D = 6.67 mm each. Each nozzle has a fuel nozzle of d = 1 mm inner diameter located upstream in tandem, such that the air and fuel mix in a channel of length \approx 5D/33 d before entering the combustion chamber. The combustor was operated with two configurations, the first was with a cylindrical quartz combustion chamber (inner diameter, I.D = 208 mm, thickness, th = 3 mm), to allow complete optical access and the second was with a stainless steel 304 combustion chamber, which had six access ports, to allow for intrusive temperature and gas composition measurements [28].

The massflow rates of CH₄, H₂ and pilot burner air were controlled by Bronkhorst El-flow flow controllers which have an accuracy of +/-0.5 % of reading plus +/-0.1 % of full scale. The main and cooling air flow rates were controlled by Bronkhorst In-Flow meters with Badger RCV Control valve leading to an accuracy of +/-1 % of full scale. The combustor was operated for fuel mixtures with H₂ % from 0 to 100 % and equivalence ratio (ϕ) varying from 0.3 to 1.0 at a constant inlet air temperature of 350 °C and constant nozzle bulk velocity of ≈100 m/s.

When operating the facility, a fixed startup procedure was followed; the system was preheated by allowing a nominal main air flow rate, set



(c) Burner head front view

Fig. 1. Combustor setup where access ports are indicated as P1-6. All dimensions are in mm.

to a temperature of 350 °C, till the temperature registered on the outer wall of the burner head was around 80 °C. At this point, the pilot burner was ignited and then the main burner was started at a nominal power of 20 kW at 100 % CH₄, ϕ =0.8. After the massflow rates and exhaust temperature stabilize, the pilot burner was turned off. The main burner power was slowly increased to 30 kW and the system was allowed to heat up until it reaches a steady state, noted by the temperature of the external wall of the burner head. Once this was achieved, the parameters were set to the desired operation conditions, and the measurements were taken once the system reached a steady state. The facility is designed to be able to operate up to a maximum of 200 kW thermal power, however, in this study, it was operated between 30 and 80 kW. Although the pilot burner was turned off, a nominal air flow of 62 lnpm was maintained to prevent damage by overheating. This served as a dilution stream in the combustor, thereby altering the effective equivalence ratio.

(b) Nozzle cross section (magnified)

Detailed settings of the operating conditions are listed in Table 1. The H₂ %, equivalence ratio (ϕ), heater temperature (set to 350 °C) and

thermal power (Q_{in}) were given as inputs to the control system. The value of ϕ specified was that for the main burner. The massflow rate set points were obtained by calculation and the values listed in the table are the average of those that were measured by the massflow controller. The temperatures listed are the average of the uncorrected measured values from Inconel sheathed K-type thermocouples.

2.2. Measurement techniques

The velocity field was measured using PIV in the quartz combustion chamber under reacting flow conditions for a mixture of $H_2 = 0$ % at a thermal power of 60 kW, $T_{air} = 350^{\circ}$ C for $\phi = 0.6$ and 0.8. TiO₂ particles of an average particle diameter of 0.5 μ m were used to seed the flow through the fuel stream in the top nozzle of the burner head, as shown in Fig. 1. The particles were illuminated with an Evergreen double pulse laser with 200 mJ pulse energy and a pulse width of 9 ns at 532 nm emission, formed into a sheet with an average thickness of

Table 1

Time averaged operating conditions.

$\rm H_2~\%$	ϕ	T _{air} (°C)	T _{fuel} (°C)	T _{mix} (°C)	\dot{m}_{air} (lnpm)	\dot{m}_{CH_4} (lnpm)	\dot{m}_{H_2} (lnpm)	U _{nozzle} (m/s)	Q _{in} (kW)	T_{cool} (°C)	T _{exhaust} (°C)	[,] m _{pilot air} (lnpm)	ṁ _{cooling air} (lnpm)
0	0.6	371	19	301	1133.7	71.5	0.0	102.3	42.6	4	519	62	2129.2
0	0.7	378	19	298	1131.5	83.2	0.0	102.4	49.6	4	547	62	2607.4
0	0.8	373	19	294	1131.3	94.8	0.0	102.7	56.6	3	566	62	3088.7
0	0.9	371	19	290	1123.0	106.4	0.0	102.4	63.5	2	581	62	3598.7
0	1	371	19	287	1121.5	117.8	0.0	102.6	70.3	2	590	62	4018.4
10	0.6	363	20	300	1133.3	69.3	7.7	102.5	42.8	4	522	62	2134.1
10	0.7	364	20	296	1127.0	80.6	8.9	102.4	49.7	4	544	62	2548.5
10	0.8	367	20	293	1123.5	91.9	10.1	102.4	56.7	4	563	62	3046.8
10	0.9	366	20	289	1122.1	103.0	11.4	102.7	63.5	3	575	62	3528.1
10	1	372	19	285	1115.9	114.1	12.6	102.5	70.4	2	592	62	4061.9
50	0.35	366	20	307	1050.0	30.8	30.8	95.2	23.9	4	452	62	1166.1
50	0.5	365	20	299	1113.7	46.9	46.9	102.0	36.4	4	496	62	1816.3
50	0.6	366	20	294	1108.5	56.0	56.0	102.2	43.5	3	528	62	2215.3
50	0.7	367	20	289	1103.7	64.9	64.9	102.4	50.4	3	548	62	2636.9
50	0.8	369	20	284	1096.8	73.8	73.8	102.5	57.3	2	564	62	3103.8
50	0.9	372	20	280	1093.0	82.5	82.5	102.8	64.1	2	578	62	3603.0
50	1	371	20	275	1086.5	91.1	91.1	102.8	70.7	2	591	62	4115.2
80	0.3	368	20	303	1110.9	17.5	70.1	102.0	23.1	5	437	62	1130.7
80	0.5	368	20	289	1090.4	28.6	114.4	102.4	37.7	4	503	62	1912.7
80	0.6	367	20	283	1077.9	34.0	135.9	102.4	44.7	4	533	62	2286.4
80	0.7	367	20	276	1064.8	39.2	156.9	102.4	51.6	3	552	62	2698.6
80	0.8	367	20	271	1054.7	44.4	177.5	102.5	58.4	3	562	62	3105.5
80	0.9	365	20	265	1048.9	49.4	197.6	103.0	65.0	3	571	62	3539.1
80	1	363	20	260	1032.4	54.3	217.3	102.6	71.5	3	575	62	3939.6
100	0.3	369	20	293	1082.6	0.0	136.9	102.0	24.6	5	436	62	1238.4
100	0.6	365	20	266	1032.3	0.0	259.6	102.9	46.7	4	535	62	2440.4
100	0.8	368	20	250	993.8	0.0	334.7	102.7	60.2	3	569	62	3316.0
100	1	369	19	237	965.4	0.0	405.0	103.2	72.8	3	582	62	4172.5

1 mm using a series of optics. Two LaVision Imager sCMOS cameras with Micro Nikkor 105 mm lenses with f# 11 were used in planar two component mode. Each camera had a pixel size of 6.5 µm and field of view 131 mm as shown in Fig. 1, denoted as FOV_{PIV}. Their frames overlapped by 24.7 mm along the axis of the combustor. Approximately 2000 images were acquired for each case at 15 Hz with a Δt of 3.5 and 6 μ s for the cases of $\phi = 0.6$ and 0.8, respectively. The uncertainty, based on Gaussian distribution, of the mean is 4 $\%\sigma_{\mu}$ (σ_{μ}/\sqrt{N}) and of the root mean square (RMS) is 3 $\%\sigma_{\mu}$ ($\sigma_{\mu}/\sqrt{2 N}$) for 95 % confidence interval, where σ_{μ} is the standard deviation of velocity. Polarisation filters were placed in front of the lens to reduce noise from reflections of the laser from the surfaces of the quartz pipe, and a 532 nm bandpass filter was used to minimize background noise and the effect of flame luminescence. The images were processed in DaVis 8.4 using an iterative multi-grid approach such that the final pass was done by a 24x24 pixel window with 75 % overlap. This resulted in a vector spacing of 305.6 μ m and a window size of 1222.4 μ m. While the resolution is relatively coarse, it is the best possible trade-off to capture the relevant field of view to observe the overall flow field.

The gas analyzer and thermocouple measurements were done using the steel chamber by inserting the probes through access ports P1-P6, as shown in Fig. 1. Each probe had a bushing such that the probe slid within the inner bore of the bushing while keeping minimal clearance. The inner bore diameter was 0.5 mm larger than the respective probe. Due to the pressure drop across the exhaust duct of the combustor, the pressure within the chamber exceeded that of the surrounding atmosphere. Consequently, gas could have leaked out of the chamber through the gap between the bushing and the probe, while the reverse flow was not possible. This condition ensured that the composition and temperature measurements were not diluted by the surrounding air. The measurements were done by traversing the probe radially at a constant speed such that a distance equal to twice the characteristic length (equal to the diameter of the probe) was traversed while acquiring 100 and 200 samples for the gas analyzer and thermocouple, respectively. The values were spatially averaged to reduce measurement noise by taking a sliding average with a convolution filter of the size of the characteristic length.

An S-type thermocouple with a platinum (10 %) sheath was used to probe the temperature within the chamber. The thermocouple measurements in the chamber were corrected using an energy balance equation,

$$\epsilon_{tc}(\sigma\epsilon_g T_g^4 + \sigma\epsilon_{wall}T_{wall}^4) + h(T_g - T_{tc}) - \sigma\epsilon_{tc}T_{tc}^4 = 0,$$
(1)

where ϵ_{ic} was the emissivity of the thermocouple sheath, ϵ_{ivall} the emissivity of the combustor inner wall, ϵ_g the emissivity of the gas around the thermocouple measurement point, T_{ic} the temperature measured by the thermocouple, T_g the temperature of the gas around the thermocouple measurement point and T_{ivall} the temperature of the inner wall of the combustor as measured by the thermocouple at r = 0.0. Details on the parameters used for the corrections can be found in Appendix B.1 in supplementary data.

A suction-based system was used to extract gas samples to measure composition in terms of NO, CO, CO₂, CH₄, and O₂. The probe was an Inconel pipe with an outer diameter of 6 mm and an inner diameter of 3 mm. The temperature just before the heated line of the gas analyzer was measured to ensure that the gases were quenched but not cold enough to allow for the condensation of water vapor. An ABB Limas21 HW process photometer (UV-RAS) (0.1 ppm accuracy) was used to measure NO under hot-wet conditions, i.e., the water vapor was not condensed out of the gas stream. This was ensured by maintaining the heated hose at 180 °C and the Limas21 module at 80 °C. The remaining gases were measured on a dry basis by condensing out the water vapor using a cooler. CO₂, CO, and CH₄ were measured using a Uras26 IR absorption analyzer (0.1 % vol/ 10 ppm/ 0.01 %vol accuracy), and O₂ was measured in the Magnos28 paramagnetic analyzer (0.1 % vol accuracy). The gas analyzer was calibrated daily using Air Liquide calibration bottles with a certified accuracy of 1 %. NO was calibrated with a bottle of NO/N2 (8.46 ppm/balance), CO and CH4 on a bottle of $\rm CO/CH_4/N_2$ (801 ppm/3.99 %vol/balance), $\rm CO_2$ on a bottle of CO2/N2 (59.8 % vol/balance) and O2 on laboratory air. The respective bottles were used for span calibration, while zero calibration was done using a bottle devoid of the species. Gas composition measurements are corrected to a standard value of 15 $\%~O_2$ using the equation:

$$\chi_{j@15\%O_2} = \chi_{j_{meas}} \left[\frac{0.2096 - \frac{15}{100}}{0.2096 - \chi_{O_2}} \right],\tag{2}$$

where χ_j is the mole fraction of species 'j' and χ_{O_2} is the measured mole fraction of oxygen.

Chemiluminescence imaging of the flame was done using a Nikon D7500 DSLR and the field of view is shown in Fig. 1, denoted as FOV_{chem}. The exposure time ranged between 100 and 800 μ s, and for $\phi < 0.4$, the value was 2.5 ms due to a fainter signal. The "blue" component of the acquired images, which has a peak around 455 nm and a full width at half max (FWHM) of ~100 nm [29], is extracted and used for further processing. This correlates with the CH* emissions (431 nm) for flames with CH₄; however, a significant blue spectrum is also seen for H₂ enriched flames up to 100 %, as was characterized by [30]. Flame kernels were identified for the instantaneous fluctuation images using an algorithm based on a combination of the Density-Based Spatial Clustering of Applications with Noise (DBSCAN) [31] and Otsu thresholding algorithm [32]. The detailed procedure is explained in Appendix B.2 in supplementary data.

3. Results and discussion

3.1. Flow field

Fig. 2a and b show the mean axial velocity field for the reacting flow at 60 kW, $H_2 = 0$ % with $\phi = 0.6$ and 0.8 respectively, to visualize the flow fields of two different nozzle bulk velocities and fields of varying levels of temperature due to the influence of ϕ . The high-velocity jet exits the nozzle and forms two recirculation zones, one towards the center of the combustion chamber, the Central Recirculation Zone (CRZ), and one towards the wall, the Peripheral Recirculation Zone (PRZ). The PRZ is a more compact region than the CRZ. The jet flow impinges on the wall at X \approx 130 mm (X/D = 19.5), closing off the PRZ, and continues as a wallbounded flow with further development of the CRZ downstream. This is a typical feature in wall offset jets parameterized by an offset ratio [33,34].

The jet entrains fluid from the recirculation zones, the extent of which is determined by tracking the inflow at the interface between the jet and the surroundings. This was done by first determining the jet centerline, which was defined as the location of maximum axial velocity for every X-coordinate. Next, the interface was detected, which was defined as the location perpendicular to the centerline where the tangential velocity changed sign. The interface and jet centerline are shown overlaid on the flowfield in Fig. 2. Finally, the velocity component perpendicular to the local interface was used to compute the entrained flow. The

local entrainment ratio (ER) ((dQ/dx)/ Q_{in} , where dQ/dx was the volume flow entrained per unit length of the interface and Q_{in} is the inlet volume flow at the nozzle) is computed for the CRZ and PRZ as shown in Fig. 3a and b. One can observe that the ER for the CRZ is high in the initial part of the chamber(up to X \approx 50 mm) after which it becomes negative, indicating a detrainment. The PRZ, on the other hand, continues to have a positive value up to X \approx 100 mm. The total recirculation ratio (RR) is computed by integrating this data along the interface and is listed in Table 2 along with the contributions from the PRZ and CRZ. Further, the PRZ contributes more than the CRZ to the recirculation ratio by a factor of 2–2.5.

This combustor has a high recirculation ratio (~2.5) on a volume basis, with a split of ~3.5 in the PRZ and \approx 1.5 in the CRZ. Combining this with the temperature measurement information presented ahead in Section 3.2, one can estimate the density ratio of the recirculated products to the incoming reactants. Estimating an average value from the measurements, the products from the PRZ are at 1250 K, from the CRZ are at 1500 K, and the inlet reactants are at 623 K. This results in a density ratio of \approx 0.5 and 0.4 for the PRZ and CRZ, respectively. Thus, the recirculation ratio on a mass basis is approximately 1.75 for the PRZ and 0.6 for the CRZ, resulting in a total RR of \approx 1.2. This would provide conditions for significant dilution of the fresh reactants and raise the temperature, making it suitable for Flameless/MILD combustion.

The turbulence intensity, shown in Fig. 4 has a similar distribution and magnitude for both cases. There is a peak value between 25 and 30 % closer to the jet exit, while further downstream in the region where recirculation begins, it reduces to around 15 %. In the downstream region beyond where the flow impinges on the wall, the value drops to 10 % and lower for $\phi = 0.6$, while it maintains a higher value, 10–15 % for $\phi = 0.8$. The lower jet velocity in the latter case may lead to less turbulence dissipation. The central recirculation zone has higher turbulence intensity than the peripheral recirculation zone due to dissipation at the wall. The high turbulence region, ≈ 15 % extends as a bulge towards the central recirculation zone in the region of 50 < X < 125 mm and -20 < Y < -40 mm.

The turbulence intensity obtained from PIV is a guideline for estimating the magnitudes under varying operating conditions. The two critical regions where reactive mixtures may be formed are at the interface of the jet and recirculation zones close to the jet exit (T.I=25 %) and further downstream, where the jet slows down and reverses to recirculate (T.I=15 %). These values are used to plot the operating conditions on the Borghi diagram and predict the behavior of premixed flames under varying inlet composition conditions. The laminar flame speed was calculated using the FreeFlame model in Cantera [35] for a mixture consisting of 50 % fresh reactants, by mass, and the remainder being burnt products. The POLIMI_C1C3_HT_NO_x_1412 chemical kinetic mechanism [36,37] was used (referred to as C1C3 in this article),



Fig. 2. Axial velocity contour map with vectors overlayed for $H_2 = 0$ %, P = 60 kW, $(a)\phi = 0.6$ (b) $\phi = 0.8$.



Fig. 3. Entrainment ratio along the combustor axis at the CRZ and PRZ for $H_2 = 0$ %, P = 60 kW, $(a)\phi = 0.6$ $(b)\phi = 0.8$.

 Table 2

 Recirculation ratio and contributions from PRZ and CRZ.

Power (kW)	ϕ	Recirculation Ratio (RR)	PRZ RR	CRZ RR
60	0.8	2.56	3.68	1.49
60	0.6	2.52	3.45	1.6

which is a detailed mechanism of the pyrolysis, partial oxidation, and combustion of hydrocarbon fuels up to 3 C atoms with NO_y. It accounts for 115 species and 2141 reactions. Sabia et al. [38] showed that the C1C3 mechanism (also called the Ranzi mechanism in the literature [36,37]) captured the low-temperature chemistry and Negative Temperature Coefficient (NTC) region of methane-air combustion the best amongst the openly available mechanisms compared in the study. As this study encompasses methane-hydrogen admixtures under vitiated conditions, requiring ignition at moderate temperatures, this mechanism was chosen. The fresh reactants were initiated at a temperature of 623 K and the burnt products were calculated for the fresh reactant mixture using a Perfectly Stirred Reactor (PSR) model. The operating conditions at various turbulence levels are plotted in Fig. 5, where it can be seen that most of the points lie in the Broadened Preheat Thin Reaction Zone (BPTRZ) region. Increasing the percentage of H₂ seems to shift the point towards the bottom right, i.e., more corrugated flamelets. Increasing the turbulence intensity from 15 % to 25 % pushes the conditions into Broadened Reactions (BR), although this is achieved only under the leanest equivalence ratio conditions with $H_2 = 50$ % and 80 %.

The case of 0 % H₂ could reach a minimum of ϕ = 0.5, below which a converged solution was not possible.

3.2. Temperature measurements

The temperature within the combustion chamber was measured by traversing an S-type thermocouple radially through access ports on the wall of the steel chamber. Temperature measurements vs radial location are shown in Fig. 6, for $H_2 = 0, 50, 80$, and 100 % at Port 2, 3, 5, and 6, the locations of which are shown in Fig. 1. The radial locations are represented as the distance from the combustion chamber wall, normalized by the radius of the chamber. The measurements have been corrected for radiation and convection based on the velocity field. The vertical lines indicate the radial location of the jet centerline (-.) and interface with the recirculation zones (- -), obtained from the flow field presented in Section 3.1.

The temperature magnitude increases with equivalence ratio, as expected. The peak values increase from Port 2 to Port 3 and then decrease at Ports 5 and 6, which indicates that heat release by combustion primarily occurs up to Port 3, and then further downstream, heat loss dominates. However, for the leanest cases of each H₂ %, i.e., H₂ = 0 % ϕ = 0.6, H₂ = 80 % ϕ = 0.3, H₂ = 100 % ϕ = 0.3, this trend differs with peak values increasing from Port 2 to Port 5, indicating a more distributed heat release zone which is shifted further downstream. From the location of the peaks, it seems that the point of highest heat release is concentrated near the interface of the jet and the recirculation zones (PRZ and CRZ). For the CRZ, side, the peak is shifted into the CRZ at



Fig. 4. Turbulence intensity for $H_2 = 0$ %, P = 60 kW, (a) $\phi = 0.6$, (b) $\phi = 0.8$.



Fig. 5. H_2 admixture conditions plotted on the Borghi diagram for $u'/U_0 =$ (a) 0.15, (b) 0.25 for a jet of $U_0 = 100$ m/s. The symbols indicate H_2 percentage in the fuel mixture and colours indicate the equivalence ratio. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Port2 and moves inward towards the jet center at successive Ports. The peak near the PRZ interface is lower than that near the CRZ at Port2 except for in the case of $H_2 = 0 \% \phi = 0.6$ and $H_2 = 80 \% \phi = 0.3$. In these exceptions, the lower or almost equal level of peak near the CRZ indicates a delay in ignition on the CRZ side, possibly due to lower entrainment, as shown in Section 3.1. Further downstream, at Port 3, the peak near the PRZ vanishes as the interface line shifts closer to the wall, resulting in higher heat loss from the reaction zone. This corresponds to the region where recirculation towards the wall initiates. The flow impinges on the wall around Port 5, where the PRZ interface is no longer present, and the jet centerline is exposed to direct heat loss at the wall. At Ports 5 and 6, even the peak near the CRZ is negligible. There is a steep drop in temperature near the wall ($0 < r_{wall}/R_0 < 0.1 - 0.25$) due to heat losses. Interestingly, the temperature values remain similar for varying H_2 content. This may be due to backflow from the cooling system, which is discussed in detail in Appendix A.1 supplementary data.

3.3. Gas composition measurement

The gas composition was measured by extracting samples through a probe traversing radially through access ports on the wall of the steel chamber. This provides an overview of the spatial distribution of the reaction zone and NO formation sites. The radial distribution of CO is shown in Fig. 7 and that of NO in Fig. 8. There are vertical lines, similar to those in the temperature plots, that indicate the location of the local jet center (-.) and the interface with the PRZ and CRZ (- -), obtained from the flow field presented in Section 3.1.

The typical pathway for combustion of CH_4 is, $CH_4 \rightarrow CH_3 \rightarrow CH_2O \rightarrow HCO \rightarrow CO \rightarrow CO_2$. Thus, in this study, CO measurements are considered a marker of a progressing reaction zone for fuel mixtures ranging from 0 to 80 % H_2 . The reader can refer to Appendix C.2 in supplementary data for CH_4 and CO_2 to verify the reaction progress. For $H_2 = 0$ %, there is a high level of CO at Port 2. For $\phi = 1.0$, a single peak is seen close to the jet centerline, while $\phi = 0.6$ and 0.8 show two peaks. The two peaks in $\phi = 0.8$ are less separated than those in $\phi = 0.6$ and occur between the jet centerline and the CRZ interface. In the case of $\phi = 0.6$, one peak is between the PRZ interface and the jet centerline and the other lies within the CRZ. Further, the CO peak is higher near the CRZ than the PRZ. This biasing of the peaks may be explained based on the information on the velocity field in Section 3.1.

It is observed that the entrainment of fluid into the jet is stronger at the interface on the PRZ side in 0 < X < 100 mm than the CRZ. This results in more recirculated combustion products at elevated temperatures and containing radicals entering the jet and mixing with fresh reactants on the PRZ side, resulting in more dilution of the fresh reactants and earlier reaction progress. On the CRZ interface side, the ignition and resulting heat release reaction are slower due to less dilution with recirculated hot products, resulting in less reaction progress and higher CO. In the case of $H_2 = 0 \% \phi = 0.6$, the CO peak lying in the CRZ suggests that the process is driven by turbulent diffusion at the interface, where some fresh reactant mixes with the hot products, resulting in a low fresh mixture fraction that ignites. This is also the observation at Port 3.

At Ports 3 and 5, for $H_2 = 0 \% \phi = 0.8$ and 1.0, there is lower CO towards the chamber center, and the value increases near the wall due to a slowing down of reactions because of heat loss, as seen in the temperature profiles in Fig. 6. The CO value is comparatively low (<100 ppm) at Port 3 and 5, indicating that most heat release reactions occurred upstream. Thus, the recirculated gases can be considered to be fully burnt products. On the other hand, for $\phi = 0.6$, there is a significant CO value at Port 3, thus leading to some unburnt mixture being recirculated in the PRZ. This can also be seen through high CO values in the PRZ at Ports 2 and 3 for this case.

The 50 % and 80 % H₂ cases with $\phi = 0.6$ and 1.0 have a very low level of CO in all Ports with a single peak at Port 2 and the magnitude decreases in Port 3 and 5 successively. This indicates that the reaction initiated and progressed significantly before the axial location of Port 2. For 80 % H₂ and $\phi = 0.3$, CO has a uniform radial distribution at Port 2, and the value increases at Port 3 and decreases at Port 5, indicating a distributed reaction zone centered around Port 3. The biasing of the CO peak towards the CRZ interface at Port 2 and 3 is also seen for $\phi = 0.6$, H₂ = 50 and 80 % at Port 3 and H₂ = 80 % at Port 2. At Port 5, $\phi = 1.0$ has higher CO than $\phi = 0.6$. This may be due to the CO–CO₂ equilibrium shift in the backward direction due to the lack of excess O₂ and higher adiabatic flame temperature. CO values are higher near the wall due to flame quenching due to heat losses and decrease towards the center due to higher temperatures.

The NO radial distribution at Ports 2, 3, and 5 is shown in Fig. 8. For the case of 0 % H₂, for $\phi = 1.0$ and 0.8, there is a trough in NO at Port 2, the region within the jet. At Ports 3 and 5, the distribution increases from the wall to the chamber center. NO is also higher for higher ϕ . In the cases of 50 and 80 % H₂, the trough at Port 2 is somewhat visible for $\phi = 1.0$, but for the rest of the cases and Ports, the values are either increasing from wall to centerline or almost constant. This is opposite to the CO trend but may be expected as the temperature



Fig. 6. Temperature vs radial location at Ports 2, 3, 5 and 6. The vertical lines indicate the local jet center (-.), PRZ, and CRZ (- -) on the left and right of the jet centerline, respectively.

increases towards the centerline. However, the temperature does not rise monotonically and has peaks around the edge of the jet, but the NO profile does not respond accordingly. This may be due to low oxygen levels in the recirculated products and the slow NO formation rate in a fast-moving flow.

The NO, measured at the exhaust of the quartz chamber configuration and compared for all operational conditions, is shown in Fig. 9a. Ultra-low levels of NO (< 5 ppm) were achieved at extremely lean conditions (ϕ < 0.6), and these conditions were only achievable, i.e., possible to stabilize in the chamber, with higher levels of H₂ in the fuel mixture. However, for H₂ = 50 % ϕ <0.4, the corresponding CO levels are high ≈1000 ppm, as shown in Fig. 9b. This indicates that for these conditions, a lower jet velocity would be preferred to increase the residence time for CO consumption with possibly minimal impact on NO, which is already ultra-low. NO increases with ϕ and seems generally higher for higher H₂ content in the fuel for the same ϕ . One can also observe that the case of 80 % H₂ goes off trend and does show higher NO than the 100 % case at ϕ =0.3 while having lower NO than the 50 % case at $\phi = 1.0$. If one observes the adiabatic flame temperature of these fuel mixtures in Table 1, the value is higher for higher H₂ content. However, the difference is minimal, i.e., $\langle = 100 \text{ K}$. Thus, other factors may be responsible for the NO trend with respect to H₂ %. CO levels at the exhaust increase with equivalence ratio and also increase suddenly at $\phi < 0.4$. The addition of H₂ decreases CO for the same ϕ .

The temperature throughout most radial locations is less than 1800 K, avoiding the typical temperature range of thermal NO_x formation. NO of a similar order of magnitude in the outer and inner recirculation regions may also indicate that although the thermal pathway is subdued, other pathways of NO_x , such as N_2O and NNH pathways, may play an important role, such that they are more dependent on gas composition than temperature. In all of the NO formation pathways, H and OH radicals play an essential role, as shown in the following equations:

$$N + OH \rightleftharpoons NO + H$$
 (3)

Ν



Fig. 7. CO mole fraction vs radial location at Ports 2, 3, and 5. The vertical lines indicate the local jet center (-.), PRZ, and CRZ (- - on the left and right of the jet centerline, respectively).

 $N_2 O + H \rightleftharpoons N O + N H \tag{4}$

 $H + N_2(+M) \rightleftharpoons NNH(+M) \tag{5a}$

$$NNH + O \rightleftharpoons NH + NO$$
 (5b)

In the thermal pathway, OH is responsible for the final production of NO (Eq. (3). In the N₂O pathway, the H radical leads to conversion of N₂O to NO (Eq. (4)). In the NNH pathway, H radical leads to NNH formation which is later oxidized to NO (Eq. (5)). Thus, the increasing concentration of H/OH radicals would aid in NO production, which would be the case for higher H₂ % in the fuel mixture.

3.4. DSLR results

Fig. 10 shows the average flame chemiluminescence, where the columns represent the variation of H₂ %, from 0 to 80 % in the fuel mixture, while the rows represent different equivalence ratios. The ultralean cases of H₂ = 50 and 80 % are shown in Fig. 11a and b, respectively, and the case of H₂ = 100 % is shown in Appendix C.4 in supplementary data. The field of view along the X axis ranges from the burner head up to X = 300 mm, whereas the Y axis ranges over almost the entire diameter of the chamber, leaving out some regions near the wall. For H₂ = 0 and 10 %, as ϕ increases from 0.6 to 1.0, the flame zone center

shifts upstream towards the burner head, from X≈150 mm to X≈90 mm and transitions from spatially distributed to compact. Similarly, increasing H₂ content to 50/80/100 % makes the flame more compact and stabilizes upstream at X≈25 mm. For higher H₂ content, 50/80/100 %, the average flame zone is less sensitive to the equivalence ratio due to enhanced reaction rates, which also leads to the flame attaching to the nozzle. The images towards the lean limits, i.e., $\phi = 0.6$ at H₂ = 0 % (Fig. 10), $\phi = 0.35$ at H₂ = 50 % (Fig. 11a) and $\phi = 0.3$ at H₂ = 80 % (Fig. 11b), have the most visually distributed regime. For each of these cases of H₂ %, the reaction zone could not be stabilized within the chamber for a lower ϕ at the same nozzle bulk velocity. The case of $\phi = 0.3$ and H₂ = 80 % shows a dual flame nature with a concentrated flame region closer to the nozzle and a distributed flame region further downstream in the chamber. This data shows that the addition of H₂ can enable the stabilization of a reaction zone at significantly leaner equivalence ratios.

3.4.1. Flame kernel statistics

The statistical behavior of instantaneous ignition kernels is captured by detecting clusters in the variance images as explained in Section 2.2. The characteristics of these clusters are quantified by assembling probability distribution functions (PDFs) for specific parameters, collected over clustered samples from ~200 images. The 100 % H₂ PDFs are not constructed as the signal-to-noise ratio of the variance images was too low to identify kernels from the background. The first parameter



Fig. 8. NO mole fraction vs radial location at Ports 2, 3 and 5. The vertical lines indicate the local jet center (-.), PRZ, and CRZ (- -) on the left and right of the jet centerline, respectively.



Fig. 9. NO and CO measured at the exhaust of combustor with the quartz chamber configuration

is the aspect ratio (AR) of the smallest area rectangle enclosing a cluster. Larger AR indicates longer, elliptic flame kernels, while smaller AR indicates more circular ones. Fig. 12a shows a comparison of PDFs for $H_2 = 10$ % for varying equivalence ratios. Increasing ϕ leads to a shift in the PDF peak from lower aspect ratios to higher values. Further, the PDF tails are also extended

Hydrogen %



Fig. 10. Average normalized chemiluminescence images obtained from the blue component of Nikon D7500 RGB sensor. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Fig. 11. Average normalized chemiluminescence images for $H_2 = (a) 50 \% (b) 80 \%$. Colorscale same as in Fig. 10.

to much higher values. Fig. 12b compares PDFs at $\phi = 0.6$ for varying H₂ % along with two ultra-lean cases, which have an in-fill under the curve. Increasing H₂ % leads to a clear increase in aspect ratio with a prominent extension of the tails. In the H₂ = 50 % case, the leanest condition of $\phi = 0.35$ shows a compact PDF with a peak around 1.75, which is lower than the location of the peak for $\phi = 0.6$, indicating a shift to rounder flame kernels. In the H₂ = 80 %, the leanest condition of $\phi = 0.3$ sows a distributed PDF with two peaks, one around AR = 2 and the other at AR = 6. There is certainly a shift in structure from the $\phi = 0.6$ case, but it also corroborates well with the observation of a dual flame structure in the mean image in Fig. 11b. The AR PDF shows that this case has two

types of flame kernels and thus indicates a transition case of stabilization mechanism.

Next, PDFs of the axial location of the center of mass of a cluster (X_{COM}) are extracted. This provides information on the probable location of flame kernels along the axial direction of the combustor. A lower value of X_{COM} means that the kernels are stabilizing in the initial regions of the jet where there is a high mean axial velocity and high turbulent fluctuation, and this is only possible if these mixtures have a high consumption speed or low autoignition delay time. This can be ascertained as the inlet velocity of the nozzle was kept constant across cases. Fig. 13a compares the PDFs for $H_2 = 10$ % at varying ϕ . Reducing ϕ leads



Fig. 12. PDF of aspect ratio of flame kernels at (a) $H_2 = 10\%$ (b) varying $H_2\%$ at $\phi = 0.6$ and ultra-lean conditions ($\phi < 0.5$).



Fig. 13. PDF of x location of Center of Mass(COM) of flame kernels at (a) $H_2 = 10\%$ (b) varying $H_2\%$ at $\phi = 0.6$ and ultra-lean conditions ($\phi < 0.5$).

to a clear shift in the peak towards a larger X_{COM} . For $\phi = 1.0$, the peak occurs at $X_{COM} \approx 40$ mm; for the leanest case of $\phi = 0.6$, it moves towards 110 mm. Thus, correlating with the flow field, the higher ϕ cases have kernels distributed in a region dominated by a recirculation inflow from the PRZ and where the jet has a "belly" towards the centerline. The leaner cases stabilize kernels downstream in the region where the jet deviates towards the wall, and there is an outflow into the PRZ from the jet. Thus, the reactants may already be mixed with the recirculated gases in the lean cases and comprise a diluted mixture.

Fig. 13b compares the PDFs at $\phi = 0.6$ for varying H₂ % along with two ultra-lean cases, which have an in-fill under the curve. For constant ϕ , there are clearly two flame stabilization regions. Higher H₂ % cases (50/80 %) stabilize upstream indicated by lower X_{COM} (≈25 mm) while lower H₂ % cases stabilize downstream(≈100–125 mm) with a wider distribution, indicating a wider spatial region of flame kernel occurrence. The ultra-lean cases, which occur at higher H₂ %, show separate characteristics. While the H₂ = 80 %, $\phi = 0.3$ case shows a concentrated PDF peak at lower X_{COM} with a long tail having significant area under the curve, the H₂ = 50 %, $\phi = 0.35$ shows a peak at around 110–140 mm with a wide distribution similar to the low H₂ % cases.

3.4.2. Flame stabilization mechanism

The recirculating hot products serve as an "igniter" for the fresh reactant jet stream, as shown in Fig. 14. They interact at a turbulentturbulent interface. Suppose the gases from the two streams mix completely before igniting. In that case, it leads to the formation of an autoignition kernel that would evolve evenly in all directions, thereby leading to a low aspect ratio (\approx 1) flame kernel. On the other hand, if the ignition occurs by diffusion of heat and radicals across the interface, i.e., flame propagation, the mixture ignites before complete mixing, leading to a flame kernel that would align with the interface, leading to longer, larger aspect ratio kernels. From the observed results, cases with lower AR tend to have larger X_{COM} and are located near the region where recirculation outflow from the jet begins. In comparison, higher AR are located more upstream, where there is an influx of recirculating gases. Thus, although not a sufficient condition, larger aspect ratio flame kernels indicate a higher probability of flame propagation, while lower aspect ratio kernels indicate autoignition stabilization. Thus, H₂ addition shifts the CH₄ flame from distributed and autoignitive to concentrated, flame propagation dominated stabilized. This is also consistent with Fig. 5, where the flame regimes are indicated on the Borghi diagram,



Fig. 14. Two types of ignition mechanisms

where the mixtures tending to flame propagation lie in the lower limits of the BPTRZ region while the autoignition ones lie closer to the WSR/Broadened Reaction region. The case of $H_2 = 80 \%, \phi = 0.3$ presents a dual nature, having both flame propagation and autoignition play significant roles in the stabilization. Thus, it represents a transition case where a further reduction in equivalence ratio would push it completely to autoignition stabilization, and an increase in $H_2 \%$ would push it towards flame propagation.

4. Conclusion

The stabilization of the reaction zone and ensuing emissions in a premixed jet-stabilized combustor were investigated for CH_4-H_2 fuel admixtures. The combustor was operated at constant jet velocity, and air preheat temperature while varying the equivalence ratio (ϕ) and H_2 content in the fuel mixture from 0 to 100 %. PIV measurements were done to measure the flowfield; the temperature was measured using an S-type thermocouple, the gas composition was measured by a suction probe attached to a gas analyzer unit, and chemiluminescence imaging was done using a DSLR camera to image the average flame as well as build PDFs of the fluctuating flame kernels.

This paper shows results on the flow field and reaction zone characteristics which can be used as guidelines by engineers when designing a jet-stabilized combustor. The results show that this combustor can stabilize a wide range of CH_4 – H_2 mixtures, facilitating fuel-flexible operations. However, to obtain minimum NO_x and CO emissions, the equivalence ratio for each H_2 % needs to be specifically chosen. A higher H_2 % allowed for stabilizing a leaner fuel–air mixture before lean blowout occurred. Further analysis was done to identify the flame stabilization mechanisms involved in this combustor based on flame kernel properties, which were shown to vary with H_2 % and ϕ . The following are the main conclusions:

- (i) The measured flow field of this combustor reveals a jet interacting with a Peripheral and Central Recirculation Zone (PRZ and CRZ) with an overall recirculation ratio estimated to be \sim 1.2 on a mass basis. The PRZ recirculation by volume is about 2 times that of the CRZ. There is a higher entrainment on the PRZ side than on the CRZ side, which leads to a different rate of reaction progress on the two sides of the jet due to variation in the dilution of fresh reactants in the jet by hot products, as is seen in the CO and temperature distributions.
- (ii) The heat loss from the walls influences the temperature and gas composition profiles. Quenching of the flame at the wall leads to higher CO and lower temperature in the PRZ.

- (iii) H₂ % and ϕ had a significant effect on the flame stabilization location and mechanism.
 - (a) Theoretically, by plotting the operating conditions on the Borghi diagram, it is seen that most conditions lie in the Broadened Preheat Thin Reaction Zone regime. The addition of H_2 pushes the conditions towards the corrugated flamelet regime while decreasing equivalence ratio pushes them towards the Well Stirred Reactor/Broadened reaction regime. However, for successful stabilization of very lean mixtures, a higher H_2 % is required.
 - (b) Chemiluminescence imaging reveals the average flame positions, where the addition of H_2 pushes the flame upstream, towards the nozzle, and makes it compact, while a leaner equivalence ratio leads to one stabilized downstream in a distributed manner.
 - (c) The ignition mechanism is interpreted from the probability distributions of aspect ratio (AR) and axial location (X_{COM}) of the flame kernels identified from the chemiluminescence images. Higher aspect ratio kernels stabilizing upstream are seen for higher H₂ content and higher equivalence ratios, indicating flame propagation dominant stabilization. On the other hand, lower aspect ratio kernels are seen for lower H₂ content and lower equivalence ratios, indicating an autoignition dominant stabilization mechanism.
- (iv) Effect of H_2 % and ϕ on emissions:
 - (a) NO levels increase with increasing H_2 in the fuel mixture for the same ϕ . A higher H_2 % leads to a higher concentration of H/OH radicals that enhance the N₂O/NNH pathway and play a role in the final steps of the thermal pathway. Further, longer effective residence time of hot gases due to upstream stabilization of the reaction zone for higher H_2 content leads to higher NO. However, increasing H_2 % allows for the stabilization of a leaner fuel–air mixture, which leads to lower NO at the exhaust.
 - (b) Distributed reaction zones are formed at the leanest conditions of each H_2 % case. For $\phi < 0.5$, this leads to ultra-low NO but also high values of CO, even at Port 5 and the exhaust. These cases also tend to have flatter/distributed temperature profiles. In these cases, operating at lower jet velocities might be more suitable for providing a longer residence time to reduce CO.

CRediT authorship contribution statement

Rishikesh Sampat: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation,

Conceptualization. **Niek Goselink:** Writing – review & editing, Investigation, Data curation. **Kaushal Dave:** Writing – review & editing, Investigation. **Ferry Schrijer:** Writing – review & editing, Supervision. **Arvind Gangoli Rao:** Writing – review & editing, Supervision, Funding acquisition.

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Declaration of competing interest

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Supplementary data

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Data availability

Data will be made available on request.

References

- International Energy Agency, The Future of Hydrogen. Technical report. Paris: IEA; 2019.
- [2] Staffell I, Scamman D, Velazquez Abad A, Balcombe P, Dodds PE, Ekins P, et al. The role of hydrogen and fuel cells in the global energy system. Energy Environ Sci 2019;12:463–91.
- [3] Huang Y, Yang V. Dynamics and stability of lean-premixed swirl-stabilized combustion. Prog Energy Combust Sci 2009;35:293–364.
- [4] Göckeler K, Krüger O, Oliver Paschereit C. Laminar burning velocities and emissions of hydrogen-methane-air-steam mixtures. J Eng Gas Turbines Power 2014;136:031508.
- [5] International Renewable Energy Agency. Green hydrogen cost reduction: scaling up electrolysers to meet the 1.5 °C climate goal. Technical report. Abu Dhabi: IRENA; 2020.
- [6] Perpignan AA, Gangoli Rao A, Roekaerts DJ. Flameless combustion and its potential towards gas turbines. Prog Energy Combust Sci 2018;69:28–62.
- [7] Gordon RL, Masri AR, Pope SB, Goldin GM. A numerical study of auto-ignition in turbulent lifted flames issuing into a vitiated co-flow. Combust Theor Model 2007;11:351–76.
- [8] Gordon RL, Masri AR, Pope SB, Goldin GM. Transport budgets in turbulent lifted flames of methane autoigniting in a vitiated co-flow. Combust Flame 2007;151:495–511.
- [9] Lückerath R, Meier W, Aigner M. FLOX combustion at high pressure with different fuel compositions. J Eng Gas Turbines Power 2008;130:011505.
- [10] Sadanandan R, Lückerath R, Meier W, Wahl C. Flame characteristics and emissions in flameless combustion under gas turbine relevant conditions. J Propul Power 2011;27:970–80.

- [11] Lammel O, Stöhr M, Kutne P, Dem C, Meier W, Aigner M. Experimental analysis of confined jet flames by laser measurement techniques. J Eng Gas Turbines Power 2012;134:041506.
- [12] Wünning J. Flameless oxidation to reduce thermal no-formation, Prog Energy Combust Sci 1997;23:81–94.
- [13] Cavaliere A, de Joannon M. Mild combustion, Prog Energy Combust Sci 2004;30:329–66.
- [14] Severin M, Lammel O, Meier W. Laser diagnostic investigation of a confined premixed turbulent jet flame stabilized by recirculation. Combust Flame 2022;243:112061.
- [15] Liu Z, Xiong Y, Zhu Z, Zhang Z, Liu Y. Effects of hydrogen addition on combustion characteristics of a methane fueled MILD model combustor. Int J Hydrog Energy 2022;47:16309–20.
- [16] Veríssimo AS, Rocha AMA, Costa M. Operational, combustion, and emission characteristics of a small-scale combustor. Energy Fuels 2011;25:2469–80.
- [17] Veríssimo A, Rocha A, Costa M. Experimental study on the influence of the thermal input on the reaction zone under flameless oxidation conditions. Fuel Process Technol 2013;106:423–28.
- [18] Veríssimo A, Rocha A, Costa M. Importance of the inlet air velocity on the establishment of flameless combustion in a laboratory combustor. Exp Therm Fluid Sci 2013;44:75–81.
- [19] Veríssimo AS, Rocha AMA, Coelho PJ, Costa M. Experimental and numerical investigation of the influence of the air preheating temperature on the performance of a small-scale mild combustor. Combust Sci Technol 2015;187:1724–41.
- [20] Ayoub M, Rottier C, Carpentier S, Villermaux C, Boukhalfa AM, Honoré D. An experimental study of mild flameless combustion of methane/hydrogen mixtures. Int J Hydrog Energy 2012;37:6912–21.
- [21] Ferrarotti M, De Paepe W, Parente A. Reactive structures and NOx emissions of methane/hydrogen mixtures in flameless combustion. Int J Hydrog Energy 2021;46:34018–45.
- [22] Medwell PR, Kalt PAM, Dally BB. Imaging of diluted turbulent ethylene flames stabilized on a jet in hot coflow (JHC) burner. Combust Flame 2008;152:100–13.
- [23] Gordon RL, Masri AR, Mastorakos E. Simultaneous Rayleigh temperature, OHand CH2O-LIF imaging of methane jets in a vitiated coflow. Combust Flame 2008;155:181–95.
- [24] Oldenhof E, Tummers MJ, Veen EHV, Roekaerts DJEM. Ignition kernel formation and lift-off behaviour of jet-in-hot-coflow flames. Combust Flame 2010;157:1167–78.
- [25] Oldenhof E, Tummers MJ, van Veen EH, Roekaerts DJEM. Role of entrainment in the stabilisation of jet-in-hot-coflow flames. Combust Flame 2011;158:1553–63.
- [26] Schulz O, Jaravel T, Poinsot T, Cuenot B, Noiray N. A criterion to distinguish autoignition and propagation applied to a lifted methane-air jet flame. Proc Combust Inst 2017;36:1637–44.
- [27] Aditya K, Gruber A, Xu C, Lu T, Krisman A, Bothien MR, et al. Direct numerical simulation of flame stabilization assisted by autoignition in a reheat gas turbine combustor. Proc Combust Inst 2019;37:2635–42.
- [28] Sampat R, Goselink N, Schrijer F, Gangoli Rao A. Operating characteristics of a flameless combustor obtained by experiments informed modelling. In: Turbo Expo: Power for Land, Sea, and Air; 2022. p. V03AT04A051.
- [29] Skorka O, Kane P, Ispasoiu R. Color correction for RGB sensors with dual-band filters for In- cabin imaging applications. Electron Imaging 2019;31:1–8.
- [30] Schefer RW, Kulatilaka WD, Patterson BD, Settersten TB. Visible emission of hydrogen flames. Combust Flame 2009;156:1234–41.
- [31] Schubert E, Sander J, Ester M, Kriegel HP, Xu X. DBSCAN revisited, revisited: why and how you should (still) use DBSCAN. ACM Trans Datab Syst 2017;42.
- [32] Otsu N, Smith PL, Reid DB, Environment C, Palo L, Alto P, et al. Otsu 1979 otsu method. In: IEEE Transactions on Systems, Man, and Cybernetics C; 1979. p. 62–66.
- [33] Miozzi M, Lalli F, Romano GP. Experimental investigation of a free-surface turbulent jet with coanda effect. Exp Fluids 2010;49:341–53.
- [34] Agelin-Chaab M, Tachie MF. Characteristics and structure of turbulent 3D offset jets. Int J Heat Fluid Flow 2011;32:608–20.
- [35] Goodwin DG, Speth RL, Moffat HK, Weber BW. Cantera: an object-oriented software toolkit for chemical kinetics. Thermodyn Transp Process 2021; https://www. cantera.org
- [36] Ranzi E, Cavallotti C, Cuoci A, Frassoldati A, Pelucchi M, Faravelli T. New reaction classes in the kinetic modeling of low temperature oxidation of n-alkanes. Combust Flame 2015;162:1679–91.
- [37] Bagheri G, Ranzi E, Pelucchi M, Parente A, Frassoldati A, Faravelli T. Comprehensive kinetic study of combustion technologies for low environmental impact: MILD and OXY-fuel combustion of methane. Combust Flame 2020;212:142–55.
- [38] Sabia P, Joannon MD, Picarelli A, Chinnici A, Ragucci R. Modeling negative temperature coefficient region in methane oxidation. Fuel 2012;91:238–45.