

Experimental Investigation of Lean Premixed Syngas Combustion at Gas Turbine Relevant Conditions: Lean Blow Out Limits, Emissions and Turbulent Flame Speed

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1. Abstract

Lean premixed combustion is considered the state-of-the art technology applied in stationary gas turbines for highly efficient, low-emission power generation using natural gas. Due to the increased interest in the integration of power generation with gasification processes, to CO₂ mitigation issues and use of solid fuels, fundamental combustion properties of upcoming new synthetic fuels (syngas) must be investigated.

This work presents some of the challenges modern low NO_x GTs are going to face when dealing with these new fuels, describing chemical and physical properties of various syngas mixtures and their operational parameters compared with a natural gas reference.

Description of operational window in terms of lean blow out is provided at gas turbine relevant conditions. Experiments were done for preheating temperature up to 772 K, pressure up to 15 bar and different inlet velocities.

Focus of this paper is also on the turbulent flame speed (S_T), analyzed with an experimental approach. taken (flame surface area derived from laser induced fluorescence data (OH-LIF)) S_T values can be calculated based on a mass continuity approach and thus represent global consumption rates.

2. Introduction

The importance of robust, low emission gas turbine combustion systems for highly reactive, hydrogen-containing fuels (e.g. syngas) is rapidly growing. This is due to increased interest in the integration of power generation with gasification processes, for the utilization of biomass, coal or tars. These integrated power systems offer the potential for reduced CO₂ emissions from power generation if combined with fuel gas decarbonization [1].

Nowadays, lean premixed combustion is the favored method for low emission power generation from natural gas used in stationary gas turbines. Homogenous mixing of the fuel and air combined with ultra-lean operation provide a reduction of flame temperature and are key to minimizing NO_x formation. Continued environmental concerns will lead to even stronger restrictions on NO_x emissions from gas turbines, prompting a need to exploit this combustion technique to its maximum potential.

	Mixture	Vol %	Simulated Process
1	H ₂ -CO-CH ₄	20-20-60	natural gas / cofiring
2	H ₂ -CO	50-50	oil gasification
3	H ₂ -CO	33-67	coal gasification
4	H ₂ -CO-N ₂	40-40-20	biomass / air blown gasification

Table 1: Syngas Mixtures

Syngas can be understood as a fuel mixture containing hydrogen (H₂), carbon monoxide (CO) and diluents such as nitrogen (N₂), whose chemical composition varies according to the specific feedstock (e.g. biomass, coal, tar) and gasification process. The combustion properties of syngases are mainly determined by their H₂ and CO contents and are generally characterized by very high (laminar) flame speeds, a wide range of flammability limits and low ignition delay times [2] which all contribute to a high risk of Flashback phenomena. H₂ distinguishes itself from CH₄ (the main constituent of natural gas) in that its flame speed is an order of magnitude higher, and its density is an order of magnitude smaller. Low emission combustion of syngas fuels is thus challenging due to the difficulty in achieving sufficient mixing before the onset of combustion. Additionally, the risk of flashback and associated overheating of the burner components places tight constraints on burner design.

Due to the peculiarity of this kind of fuels, syngas combustion is mostly adopted in diffusion flames and highly diluted (with N₂ or steam) combustion [3; 4].

In spite of the quantity of current research activity involving syngas [5-7], a deficit exists for lean premixed combustion experiments at gas turbine relevant conditions. More specifically, there is very little high-pressure data for turbulent, lean premixed syngas flames available. Changes in flame characteristics at elevated pressures (flame position, flame structure, turbulent flame speeds, pollutant formation...etc.) necessitate such experiments.

3. Experimental setup

The experiments were carried out in a generic, high pressure combustor shown in Figure 1. The combustor with complete optical access is specifically designed to study turbulent, lean premixed flames. It is capable of operation up to a pressure of 30 bars, with a maximum air flow rate of 750 m³/h (0.3 kg/s) and adiabatic flame temperatures up to 1950 K. The combustion air can be electrically preheated as high as 823 K. The combustor has a maximum

thermal power of 400 kW. The cylindrical liner consists of two coaxial quartz glass tubes (inner quartz glass tube diameter $D = 75$ mm) which are convectively air-cooled. The combustor inlet diameter d is 25 mm. The flame is stabilized by the recirculation of hot flue gases due to the sudden expansion geometry. Starting the rig a hydrogen torch igniter is used to ignite the premixed fuel/air mixture, which can be homogenized by means of up to three static mixer stages. More detailed descriptions of the test facility can be found in previous articles [8; 9].

The experimental conditions of the studied flames are presented in Table 2.

bulk velocity at inlet u_{bulk}	40 – 60 – 80
inlet temperature [K]	673
pressure [bar, absolute]	5 to 15
equivalence ratio	0.24 to 0.50

Table 2: Experimental conditions.

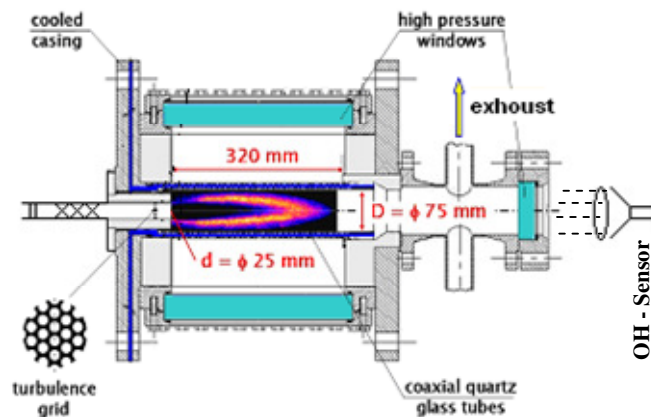


Figure 1: High-pressure combustor with OH-Chemiluminescence sensor.

Taking advantage of the optical access, a sensor was used to record the OH chemiluminescence signal at a rate of up to 1.2 kHz (see also Figure 3). Due to the sensor design and its position (Figure 1) the resulting signal is proportional to the OH chemiluminescence integrated over the whole flame [10].

LBO limits are determined using a combination of measurement techniques described below. These tests are typically carried out, starting from a stable flame at a fixed fuel to air ratio and then slowly decreasing the fuel flow rate in steps, keeping the other conditions constant, until LBO occurs. Then, towards LBO, the flame sensing signal begins to fluctuate heavily and large regions of local extinctions are observed (extended periods (few seconds) of zero intensity; see signal trace shown in Figure 2a)[9]. Proximity to lean extinction is made even more evident by a rapid increase in the CO concentration indicating incomplete combustion due to (local) extinction events. The flame then enters a self exciting mode (fluctuations are getting stronger and stronger) in which its position, shape and length change rapidly until LBO occurs. An example of the OH chemiluminescence time series of a lean blowout event is shown in Figure 2. The OH- chemiluminescence signal has been acquired with a photomultiplier using an interference filter. The sampling rate was 1200 Hz which led to a resolution of 600 Hz in the Fourier spectrum, a “time window” of one second was selected to analyze the signal (Figure 2b).

In order to measure and derive parameters such as the turbulent burning velocity, instantaneous and most probable flame position, flame brush thickness etc. an experimental campaign using planar laser induced fluorescence of OH radicals (OH-PLIF) was conducted. The setup for the OH-PLIF is described in details in [11].

For each measured operative condition 400 images (instantaneous shots) were acquired (at about 20Hz) in order to ensure a good statistic. Examples of instantaneous shots are depicted in figure 2.

The data post processing was done applying an already developed methodology [12] which has been further improved and tuned on ultra-lean syngas flames.

4. Results

4.1. Lean blow out limits

To detect the “Lean Blow Out” limits, an integrated OH-Chemiluminescence signal has been acquired by means of a high frequency photomultiplier. The obtained signal was analyzed in both time and frequency domains. This analysis led to a definition of the “Lean Blow Out” limit as the starting point of low frequency oscillations occurring as the flame approaches extinction. These oscillations are attributed to local extinction/re-ignition phenomena which can trigger pressure waves feeding into thermoacoustic feedback loops. In some cases LBO could be detected not as a direct consequence of incomplete combustion (high CO emissions), but as a result of these thermoacoustic coupling phenomena.

The LBO limit has been defined as the point in which the combustion process starts to show oscillations characterized by high amplitude and low frequency. The procedure for LBO detection is described in a more detailed manner in [13].

In Figure 2, lean stability limits (LBO) are summarized in terms of Equivalence Ratio and Pressure.

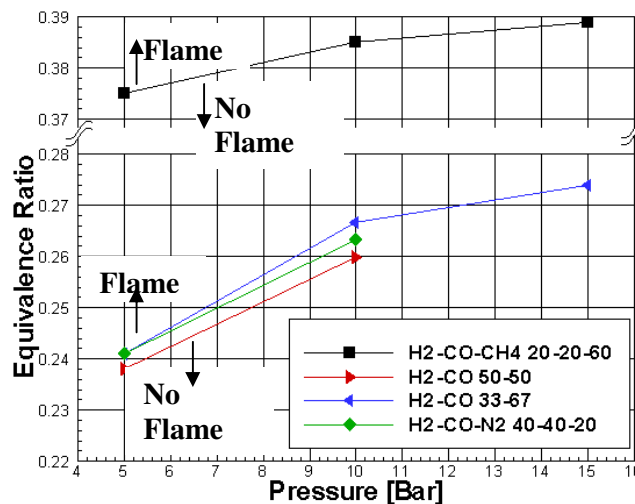


Figure 2: LBO limits for the investigated mixtures. Conditions: Inlet Velocity 40m/s, Preheating Temperature 673 K

In the Figures 3 and 4, NO_x and CO emissions are presented for all investigated mixtures at several conditions. All the acquired points are representative of a steady-state conditions unless for the last and highest CO points, in which the flame is close to extinction. Apart from extremely lean conditions (close to LBO), the measured CO emissions are always close to zero. Because of the low flame temperature at equivalence ratios close to LBO and a

higher probability of local extinction events at these conditions the CO concentration increases while approaching LBO. For these low temperature conditions the residence time is then not long enough for complete oxidation of CO.

As it is shown in Figure 2 the LBO limits were found to be almost pressure independent for the CH₄ containing mixture (Num. 1, Tab.1). All other mixtures show a slightly increased pressure dependence: the flame extinguishes at higher Equivalence Ratio with increasing the pressure. No significant dependence of the LBO limits on the inlet velocity was found for the three “pure” syngas mixtures.

Figure 3 shows the dependence of the LBO limits on the pressure for the fuel mixtures H₂-CO 33-67 (left hand side) and H₂-CO-CH₄ 20-20-60 (right hand side). The presented data were taken at 5/10/15 Bar, with preheating the mixture up to 673 K. For all “pure” syngas mixtures, the Blow Out takes place at higher equivalence ratios with increasing pressure and the CO emissions show a steeper gradient approaching LBO at high pressures. A different behavior is observed regarding the methane containing mixture (right hand side), where the LBO limit is almost at the same Equivalence Ratios for each pressure and the CO gradient approaching LBO changes only very little with increasing pressure. In the following an explanation concerning the different pressure behavior, based on the difference in CO gradients, is presented.

In Figure 3, a detail of the increasing CO emissions is shown. It can be noted that for the syngas mixture (left hand side), incomplete combustion, characterized by non-zero CO emissions, starts for all the investigated pressures (5, 10 and 15 Bar) around almost the same Equivalence Ratio of 0.285. For leaner conditions, the steep increase in CO for higher pressures indicates extinction at less lean conditions. The phenomenology behind the CO gradient can be investigated with the help of the chemiluminescence spectra. As it has been shown in [13], more energy is accumulated in the low frequency range approaching LBO which results in an oscillation characterized by a dominant frequency of 5 Hz. The energy increase can be interpreted as an evidence of the coupling between heat release oscillations (detected with the photomultiplier) and the fluctuations in the pressure field. The feedback between these two parameters is given by means of the acoustic characteristic (i.e. geometry) of the combustion chamber and the “gain” of this “closed loop” is proportional to the output power of the combustor. The increase of power with pressure thus results in a stronger amplification of the oscillations leading to quick extinction.

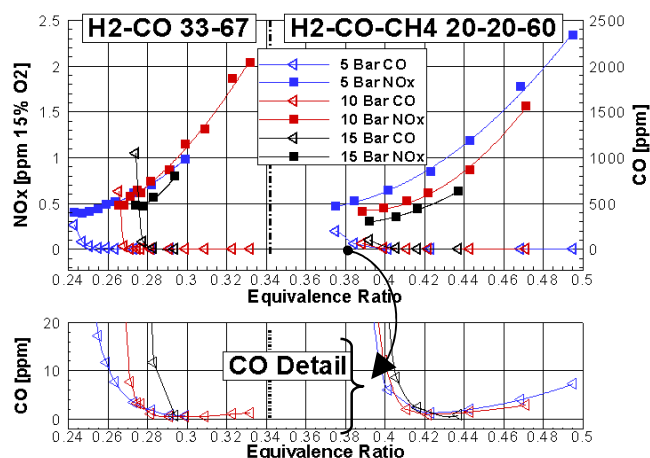


Figure 3 NO_x and CO emission: effect of the pressure on the mixture H₂-CO 33-67 on the left hand side and on the mixture H₂-CO-CH₄ on the right hand side. Conditions: Inlet velocity 40 m/s, Preheating Temperature 673 K

The described behavior is also valid for the CH₄ containing mixture (Fig.3, right hand side). Though in this case, due to slower hydrocarbon oxidation chemistry even at low pressure (5 Bar) enough time for amplification to the thermoacoustic feedback loop is given. As can be seen in Figure 3 (right hand side) the CO increases in a similar way for all pressure levels investigated.

In Figure 4, a comparison between the three “pure” syngas mixtures is presented. Looking at the CO emissions (as usual the last and highest acquired value is representative of the LBO limit), the oil-syngas mixture (Num. 2, Tab.1) shows the most extended LBO limits, although the difference with the other syngas mixtures is not very pronounced. This might be attributed to the higher H₂ content in this fuel mixture which provides higher concentrations of OH radicals and a higher laminar flame speed leading to an enhancement of the flame stability [14; 15].

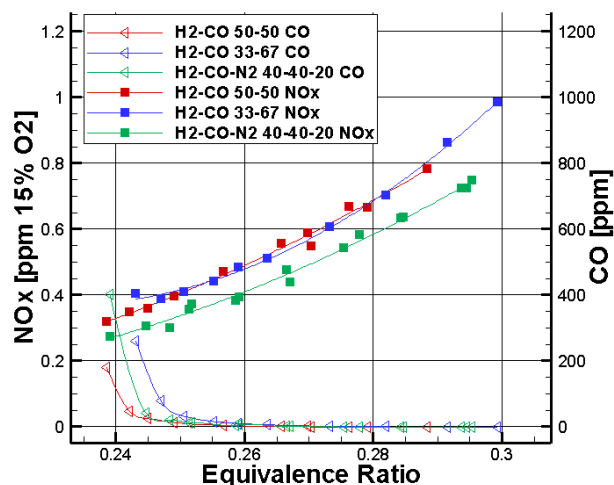


Figure 4: LBO limits and NO_x comparison between the mixtures: H₂-CO 50-50, H₂-CO 33-67, H₂-CO-N₂ 40-40-20. Conditions: Pressure 5 Bar, Inlet velocity 40 m/s, Preheating Temperature 673 K.

4.2. NO_x emissions

NO_x emissions for a wide, representative set of measuring points are presented in Figure 5c; to compare different mixtures, the results are plotted over the adiabatic flame temperature instead of the Equivalence Ratio. For sake of consistency with the previous diagrams, the latter is also displayed in the upper part of the figure (a). In the middle part (b) the O₂ percentage measured in the exhaust is also displayed.

Concerning the “pure” syngas mixtures, the upper limit in the Adiabatic Temperature was determined, by the occurrence of Flash Back. Only with low turbulence inlet conditions (no turbulence grid in fuel supply pipe; see Fig. 1) and moderate operating pressure (5 bar) the syngas mixtures could be operated up to equivalence ratios of $\Phi \approx 0.45$ corresponding to adiabatic flame temperatures around 1950K. With turbulence grid present the operational range was strongly limited ($\Phi_{\text{flashback}} \approx 0.35$). Further test are currently performed and a detailed discussion of flashback phenomena for syngas fuel mixtures will be presented in a future publication.

The measured points belonging to the “pure” syngas mixtures are contained in a narrow band which shows an exponential trend (note the logarithmic scale on the NO_x axis), according to the characteristics of thermal NO_x production. The small difference between the three mixtures could be seen more clearly in Figure 4. The N₂-diluted mixture (simulating biomass gasification), characterized by lower adiabatic flame temperatures (the difference is on the order of 30 degree), exhibits lower NO_x, while undiluted “coal & oil gasification” syngas

mixtures with slightly higher adiabatic flame temperature (for a given equivalence ratio) lead to the higher NO_x emissions.

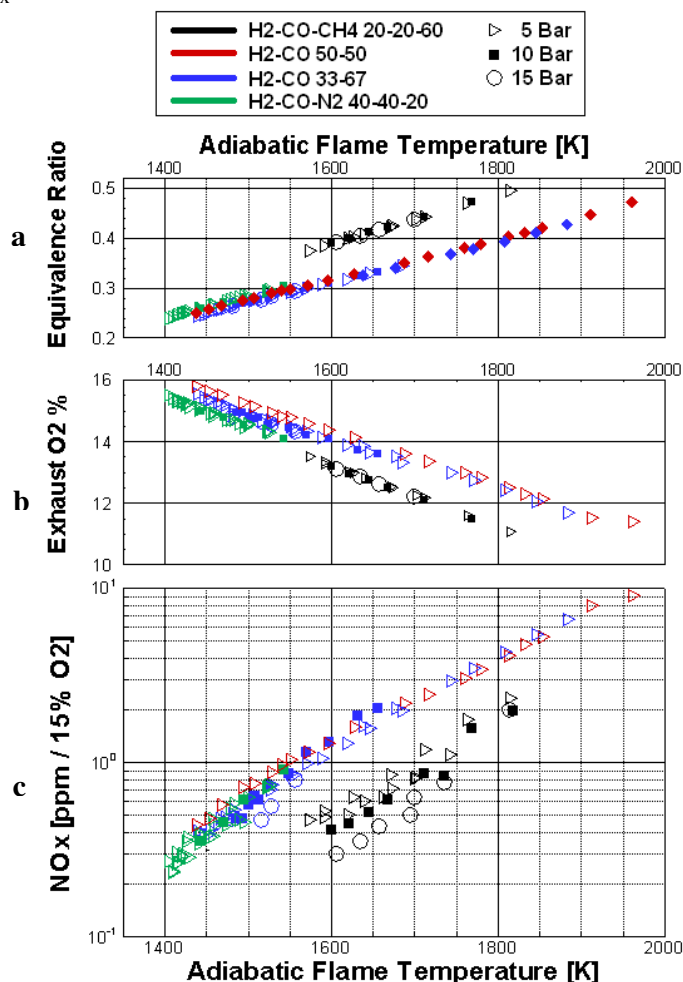


Figure 5: NO_x emission for all the investigated mixtures at several conditions

Compared to the “pure” syngas mixtures, Figure 5 shows that the data belonging to the CH₄ containing fuel mixture (simulating co-firing of syngas and natural gas) do not fall in the same range but are characterized by considerably lower NO_x emissions. This can be attributed to two different factors. First, the difference in the residence time between the flame front and the exhaust probe has to be taken in account. Figure 6 presents two OH-LIF (Laser Induced Fluorescence) images which help to visualize the distances (d_1 , d_2) leading to the different residence times in the hot exhaust flow up to the sampling point.

In Fig. 6, two flames (H₂-CO-CH₄ 20-20-60 and H₂-CO 50-50) are compared at the same adiabatic flame temperature of about 1700 K. It can be estimated, looking at the different flame lengths, that the mentioned residence time is more than doubled in the case of the H₂-CO 50-50 mixture, considering that the exhaust probe is placed at the very end of the chamber (e.g. right edge of the pictures). The difference in the residence time is estimated to be on the order of 10 ms for the given flames (corresponding to a distance d_2-d_1 of about 10-12 cm; Figure 6) considering a mean velocity of 10-12 m/s in the combustion chamber (about 30% of the inlet bulk velocity). According to chemical kinetic calculations of thermal NO_x formation this leads to a difference in the NO_x values on the order of 0.1 ppm (@ 1700 K), which partially justifies the gap in the emission data in Figure 5.

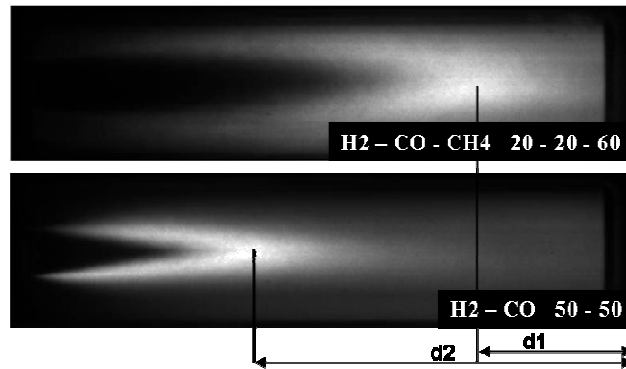


Figure 6: OH-LIF images for the two mixtures: H₂-CO-CH₄ 20-20-60 and H₂-CO 50-50. Conditions: Adiabatic Flame Temperature 1700 K, Pressure 5 Bar, Inlet velocity 40 m/s, Preheating Temperature 673 K.

The other effect to consider is the dependence of the “thermal NO” formation rate on the oxygen (O₂) concentration in the exhaust gas as described by[16]:

$$\frac{d(NO)}{dt} \propto (O_2)_{eq}^{0.5} \cdot (N_2)_{eq} \quad (1)$$

Comparing the CH₄ containing mixture with pure syngas mixtures, it has to be taken into account that for a fixed adiabatic flame temperature, the percentage of oxygen available after the flame front is considerably different (Figure 6b). E.g. at an Adiabatic flame temperature of 1700 K, the oxygen percentage is between 12Vol.% and 13.5Vol.% for co-firing syngas and for pure syngas, respectively. This difference results in a difference in the NO formation rate, according to equation 1, of about 6%, which justifies a NO_x increase again on the order of 0.1 ppm.

Only for the methane containing fuel gas mixture an effect of operating pressure on the NO_x emissions can be discerned. Higher operating pressure leads to lower NO_x emissions indicating a reduced peak concentration of radicals promoting NO_x formation (this can be due to faster recombination reactions at elevated pressure conditions). Apparently due to the different chemical kinetic pathways for hydrocarbon oxidation, only with the presence of hydrocarbons, pressure sensitive elemental reaction steps become important which influence NO_x formation via concentrations of O/H and CH_i radicals. This effect is obviously mostly limited to the main reaction zone (flame front) as the slope of NO_x emission data with respect to adiabatic flame temperature is largely maintained for all pressure conditions (indicating the general importance of thermal NO_x formation in the hot exhaust gas at all operating conditions and for all fuel mixtures).

4.3. Turbulent flame speed

The presence of H₂ and CO in the fuel mixture leads to an improved stability of the combustion process due to the faster chemistry characteristic of these fuels. Although the improved stability represent a big advantage in several operative conditions, like in case of partial load of the engine or during fast load changes, it brings with itself the very critical aspect of a strongly enhanced flashback risk. Flash Back happens when the flame propagation velocity locally overtakes the convective velocity of the incoming fuel/air mixture. The flame front starts to propagate upstream of the region where it is supposed to anchor into zones characterized by low flow velocity (shear layers) and high fuel concentration (injectors). The fuel injector and its vicinity are usually not designed to handle high temperatures and can rapidly be destroyed by the flame.

An important parameter for turbulent premixed flames which gives guidelines for combustor design engineering is the turbulent burning velocity (S_T). It provides information directly related to flashback risk, flame stability and flame length. S_T is the propagation velocity of a combustion wave in a turbulent flow field; it represents an overall property of the particular mixture adopted, operative conditions (pressure, temperature) and turbulence level all together. The corresponding parameter for laminar flow conditions is the so called laminar flame speed (S_L) which can be calculated as fundamental property of the fuel mixture upon the operative conditions. As it is not depending on the turbulence it is independent on the combustion device used.

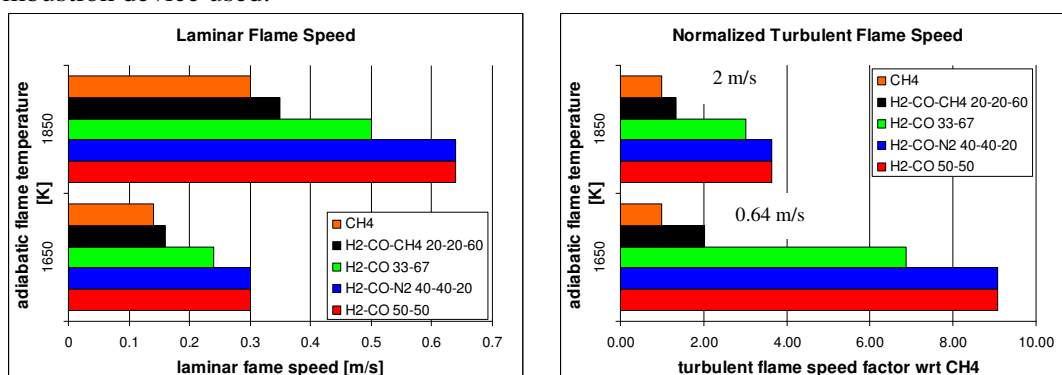


Figure 7: Laminar and Turbulent flame speeds

In Figure 7, laminar and turbulent flame speed values for four fuel mixtures at two different adiabatic flame temperatures are reported. The two temperatures were selected as representative for the lower and the upper operational limits for a conventional stationary GT engine. The S_L values were calculated with Cantera, a public domain code using a freely propagating laminar flame routine and adopting GRI3.0 as reaction mechanism; S_T values come from own experiments and are based on flame front detection via laser induced fluorescence of the OH radical [17]. Turbulent flame speeds are normalized by the corresponding CH_4 reference value. Looking at the laminar flame speed values it is possible to isolate the pure chemical kinetic properties of the different mixtures. Depending on their hydrogen-content the fuel mixtures have the ability to propagate faster and this is the case in a similar fashion for both the considered temperatures. Looking at the turbulent flame speed values the trend seen for S_L is still respected: higher H_2 fraction in the fuel mixture corresponds to a higher S_T . For low flame temperature (1650 K), though the higher H_2 content brings about additional acceleration which leads to values of almost an order of magnitude higher with respect to the corresponding value for CH_4 . This additional effect is mostly due to the strongly different physical properties (e.g. diffusivity) of H_2 and further explained and analyzed in [17].

The presented analysis highlights that the co-firing mixture shows only a moderate difference with reference to CH_4 . When compared with NG its S_T values show a factor up to 2. This fact has to be carefully taken into account when adapting burner and combustion chamber for the new fuel.

In general it can be stated that while the co-firing possibility looks achievable for a GT requiring only minor modifications, to have an engine which offers full flexibility, operating at different conditions and with undiluted syngas, represents still a major challenge.

5. Conclusions

Experimental investigation of Lean Blow Out (LBO) limits, NO_x emissions, and turbulent flame speed for lean premixed syngas/air flames in a generic combustor at conditions relevant

to low-emission, stationary gas turbines was done. The experiments were performed for a broad set of parameters including an inlet velocity variation from 40 to 80 m/s, operating pressures between 5 and 15 bar and adiabatic flame temperatures up to 1950 K.

A methodology for LBO limit detection based on the Chemiluminescence acquisition and its frequency analysis has been presented. With this approach, LBO can be detected before actual “flame out” occurs. Thus this non-intrusive method – based on the flame chemiluminescence signal captured by means of an Optical Fiber arrangement – could potentially be applied in real Gas Turbine Combustors and be used to avoid tripping of the machine with all its related negative consequences.

Not surprisingly the results show that the higher the H_2 in the fuel mixture the leaner the equivalence ratios for which LBO flame extinction occurs. The difference is significant for fuel mixtures with/without methane and can be attributed to the higher laminar flame speeds of hydrogen containing syngas mixtures (compared to natural gas/methane), even though the difference for the three “pure” syngas mixtures (Num 2, 3 and 4, Tab.1) is very small.

The NO_x emissions of all fuel mixtures investigated were found to be exponentially dependent on the adiabatic flame temperature, in line with “Thermal NO_x ” formation characteristics. “Pure” syngas mixtures have a similar behavior and their NO_x emissions fall in a narrow band. The syngas mixture with methane (number 1, Tab. 1) shows significantly lower NO_x emissions which can be partially justified with the difference in the flame length (and related residence time effects) and in the percentage of oxygen available after the flame front which distinguishes this mixture from pure syngas fuels. This fuel gas mixture with methane has found to be also slightly more sensible to Inlet Velocity and Pressure variations than pure syngases mixtures.

Understanding of the different behavior of the pure syngas mixtures involves considerations about the molecular diffusion and stretch effect that are typical for H_2 -rich mixtures. Hydrogen percentage in the fuel compounds was detected as important parameter in the determination of S_T and more specifically in the ratio S_T/S_L . The higher the H_2 in the mixture is the bigger S_T/S_L is. For low H_2 content the fuel shows a hydrocarbon-like behavior. The peculiarities of H_2 -rich flames were found to be more pronounced for ultra-lean conditions, when the fuel is the deficient species. As expected, the results highlight the strongly elevated values of turbulent flame speed for high hydrogen containing fuel gas mixtures. Compared with flame speed data for pure CH_4 the ratio ($S_T^{Syn}/S_T^{CH_4}$) takes up values of almost 10. In absolute terms values go up to well beyond 10 m/s. For ultra-lean conditions high burning velocities can be maintained with increased H_2 content in the fuel mixture.

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